An Examination of the Factors that Control Methylmercury Production and Bioaccumulation in Maryland Reservoirs

MARYLAND
DEPARTMENT OF
NATURAL RESOURCES

Resource Assessment Service

MONITORING AND
NON-TIDAL ASSESSMENT DIVISION

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An Examination of the Factors that Control Methylmercury Production and Bioaccumulation in Maryland Reservoirs

Final Report

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Summary

Human, wildlife and regulatory concerns prompted this study into the factors that affect the accumulation of methylmercury (MeHg) in fish in Maryland lakes and reservoirs. The Maryland Department of the Environment (MDE) issued fish consumption advisories for all freshwater lakes in Maryland in December, 2001. Maryland watersheds receive elevated rates of mercury (Hg) deposition, and deposition rates appear to vary substantially across the state. Watersheds across the state also have different abilities to convert inorganic mercury into MeHg, the form that accumulates in food webs. Some of these factors may be controllable through emissions regulations, or by reservoir or land use management. The overall study objective is to provide the state of Maryland with information to aid in management of MeHg bioaccumulation in fish in Maryland reservoirs.

The concentration of mercury in fish (normalized by size and species) varies substantially among Maryland lakes and reservoirs. The objective of this study was to determine which characteristics of Maryland lakes and their watersheds contribute most to this variability. An ecosystem's sensitivity to Hg loading is defined as the ability of that ecosystem to transform inorganic Hg load into MeHg in biota. Three major groups of processes contribute to that sensitivity: 1) mercury transport to zones of methylation, 2) net production of MeHg, and 3) MeHg bioaccumulation through food webs. Therefore, a broad suite of characteristics were examined, including land use in the watershed, watershed size, the physical structure of the water bodies, water and sediment chemistry, and mercury concentrations and deposition rates.

To identify those factors, we compiled a comprehensive data set on fourteen Maryland reservoirs, and used that data set to examine relationships between key variables. Data used in the analysis include newly collected (2003-2005) sediment and water samples from the reservoirs, as well as our previously collected information on water chemistry and mercury (Hg) in fish (2000-2001). The large body of cause and effect research on the controls on Hg in fish led us to choose the variables examined. Mercury deposition rates were estimated from MD DNR's recent model of wet + dry mercury deposition across the region (Sherwell et al. 2006). MD DNR's Power Plant Research Program has funded much of the research on Hg in Maryland to date. This support has resulted in assessment of the concentration and form of mercury in atmospheric deposition, in watersheds, and in biota. The data collected and compiled here builds on that foundation.

Reservoirs were chosen based on: 1) availability of Hg data from largemouth bass from our prior work, 2) representation in the major geographic provinces of Maryland, and 3) a range of land use, size and water chemistry. Western Maryland lakes were examined by Castro/Appalachian Laboratory; and

eastern lakes were examined by Gilmour/Smithsonian Environmental Research Center and Mason/Chesapeake Biological Laboratory.

Rather than a using a lumped statistical approach that examined Hg concentrations in fish against all potential controlling variables, we chose to examine each of the major steps in the Hg cycle separately. Statistical analyses of similar data sets in other regions have not been done this way. Because of the complexity of the Hg cycle, the large number of variables that affect Hg levels in fish, and the relatively small number of reservoirs examined, this approach provided more power to assess potential controls on Hg bioaccumulation.

As expected, stepwise regressions models for Hg in largemouth bass against all other variables revealed a strong correlation with MeHg levels in water, and with pH, but little more. Models for each component of the Hg cycle revealed the sequential controls on bioaccumulation.

Mercury transport from the landscape to lake sediments and bottom waters, where MeHg production occurs, is the first step in the cycle that leads to MeHg in fish. Stepwise linear regression of variables (transformed to achieve normality) showed that land use, water and sediment chemistry and Hg deposition rates explained most of the variability in Hg in sediments and water. Land use, particularly the percent of land developed, accounted for about 35% of the variability of Hg in water. One likely explanation is enhanced transport of atmospherically deposited Hg across impervious surfaces; another is direct Hg contributions from developed landscapes. Water column Hg concentrations dropped dramatically with increasing percent forested land in the watershed. The potential role of forested buffers in minimizing Hg transport to receiving waters should be investigated as a control mechanism for Hg in fish.

Importantly, Hg deposition rates explained a significant portion of the variability in water column Hg concentrations, after land use and water chemistry were accounted for. The variability in sediment Hg concentrations was driven by the grain size and organic matter content of sediments, but Hg deposition rates also contributed. These relationships support the idea that variation in mercury deposition rates across Maryland contribute to differences among lakes in fish Hg levels.

The next step in the cycle is production of MeHg. Mercury concentrations in sediment and water, along with pH, sulfate, dissolved oxygen (DO), and organic matter were the best predictors of MeHg in sediment and water. The major control on MeHg production in both sediment and water appears to be the inorganic Hg concentration. Sulfate and pH accounted for significant additional variability in water column MeHg. Sulfate stimulates MeHg production through the action of sulfate-reducing bacteria. Acidity is also commonly identified as a correlate of MeHg in aquatic ecosystems, affecting methylation, partitioning, and bioaccumulation. These relationships support the idea that reduction in acid deposition to freshwater ecosystems – particularly sulfates – will reduce the net production of MeHg from inorganic Hg. Low DO in lake bottom waters was also strongly correlated with MeHg.

The last step in the cycle is accumulation of MeHg through food webs. The bioaccumulation of MeHg from water to fish was related to DO, pH and the reservoir surface to water ratio. Reservoirs with low or zero DO bottom water had generally higher bioaccumulation factors (BAFs). Turnover of high MeHg bottom waters into surface waters in the fall may increase MeHg levels in water well above those measured in the summer. A positive relationship between BAF and surface to water area ratios suggest direct MeHg uptake from sediments.

Coastal Plain reservoirs seem particularly sensitive to Hg, in part because of low DO in bottom waters, and in part because of low pH. This DO relationship suggests a link to lake trophic status. In other ecosystems, BAFs often decrease with increasing lake productivity. However, increased rates of MeHg production in these more anaerobic systems may negate that advantage.

Summary of Recommendations

Reduce Hg emissions in Maryland. Examine any trading approaches carefully to minimize deposition hot spots in the state.

Investigate the potential role of forested buffers, porous surfaces and land use controls in minimizing Hg transport to receiving waters. There appear to be multiple negative aspects of developed landscapes on Hg cycling.

Reduce SO_X emissions in Maryland. Sulfate and pH are important drivers of MeHg production in Maryland.

Improve understanding of “dry deposition,” in order to improve understanding of total Hg deposition rates, mechanisms, sources and remediation

Adaptively manage Hg reduction strategies by developing long-term programs to monitor Hg deposition and Hg bioaccumulation across Maryland. Monitoring should begin as soon as possible so that a baseline can be established prior to implementation of new emissions regulations.

Repeat water column sampling in T.H Duckett reservoir. Anomalously high inorganic Hg levels were found in two different years.

Table of Contents

List of Tables

List of Figures

List of Abbreviations and Acronyms

AL	University of Maryland, Appalachian Laboratory
AVS	Acid-volatile Sulfides
BAF	Bioaccumulation Factor
BDL	Below Detection Limit
CALPUFF	Acronym for a numerical model for atmospheric mercury deposition, developed by the U.S. EPA
CBL	University of Maryland, Chesapeake Biological Laboratory
CRS	Chromium-Reducible Sulfides
DNR	Department of Natural Resources
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
Hg	Mercury
HgD	Filterable mercury
HgUNF	Unfiltered mercury
LMB	Largemouth Bass
LOI	Loss on Ignition
MeHg	Methylmercury
MeHgD	Filterable MeHg
MeHgUNF	Unfiltered MeHg
MDE	Maryland Department of the Environment
MDN	Mercury Deposition Network
PN	Particulate Nitrogen
PC	Particulate Carbon
PPRP	Power Plant Research Program
SERC	Smithsonian Environmental Research Center
TMDL	Total Maximum Daily Load
TSS	Total Suspended Solids
US EPA	United States Environmental Protection Agency
WHO	World Health Organization
%MeHg	(MeHg/Hg) *100

Introduction and Study Objectives

We examined the factors that may contribute to the accumulation of mercury in fish in many Maryland reservoirs. To identify those factors, we compiled a comprehensive data set on fourteen Maryland reservoirs (Figure 1, Table 1), and used that database to examine relationships between key variables. Size- and species- normalized Hg levels in fish vary by almost a factor of 10 across the 14 Maryland reservoirs examined in this study (Figure 2). Data used in the analyses include newly collected (2003-2005) sediment and water samples from the reservoirs, as well as our previously collected information on water chemistry and mercury (Hg) in fish (2000-2001). In addition, land use patterns, soil types, lake morphometry, and modeled Hg deposition rates were also examined. To examine the controls on methylmercury (MeHg) bioaccumulation, we considered the major factors that affect deposition, transport to surface waters, MeHg production and finally MeHg accumulation in food webs. Much of the research on Hg in Maryland to date has been funded under the auspices of MD DNR's PPRP program, and this work has allowed assessment of the concentration and form of mercury in atmospheric deposition, in watersheds, and in biota (Mason et al. 1997a; 1997b; 1999; Mason et al. 2000b; Gilmour, 1999; Sveinsdottir and Mason, 2003). The data collected and compiled here builds on that foundation.

Fourteen impoundments spread across the geographic provinces in Maryland were examined (Figure 1, Table 1). Reservoirs for study were chosen based on availability of Hg data from largemouth bass from our prior work, to represent the major geographic provinces of Maryland (Table 2), and to include a range of land uses (Table 3). The study included collection of sediment and water chemistry data, including Hg and MeHg concentrations, for all of these reservoirs. A variety of other physical and chemical parameters were measured simultaneously. Western Maryland lakes were examined by Castro/Appalachian Laboratory; and eastern lakes were examined by Gilmour/Smithsonian Environmental Research Center and Mason/Chesapeake Biological Laboratory. The overall study objective is to provide the state of Maryland with information to aid in management of MeHg bioaccumulation in fish in Maryland reservoirs.

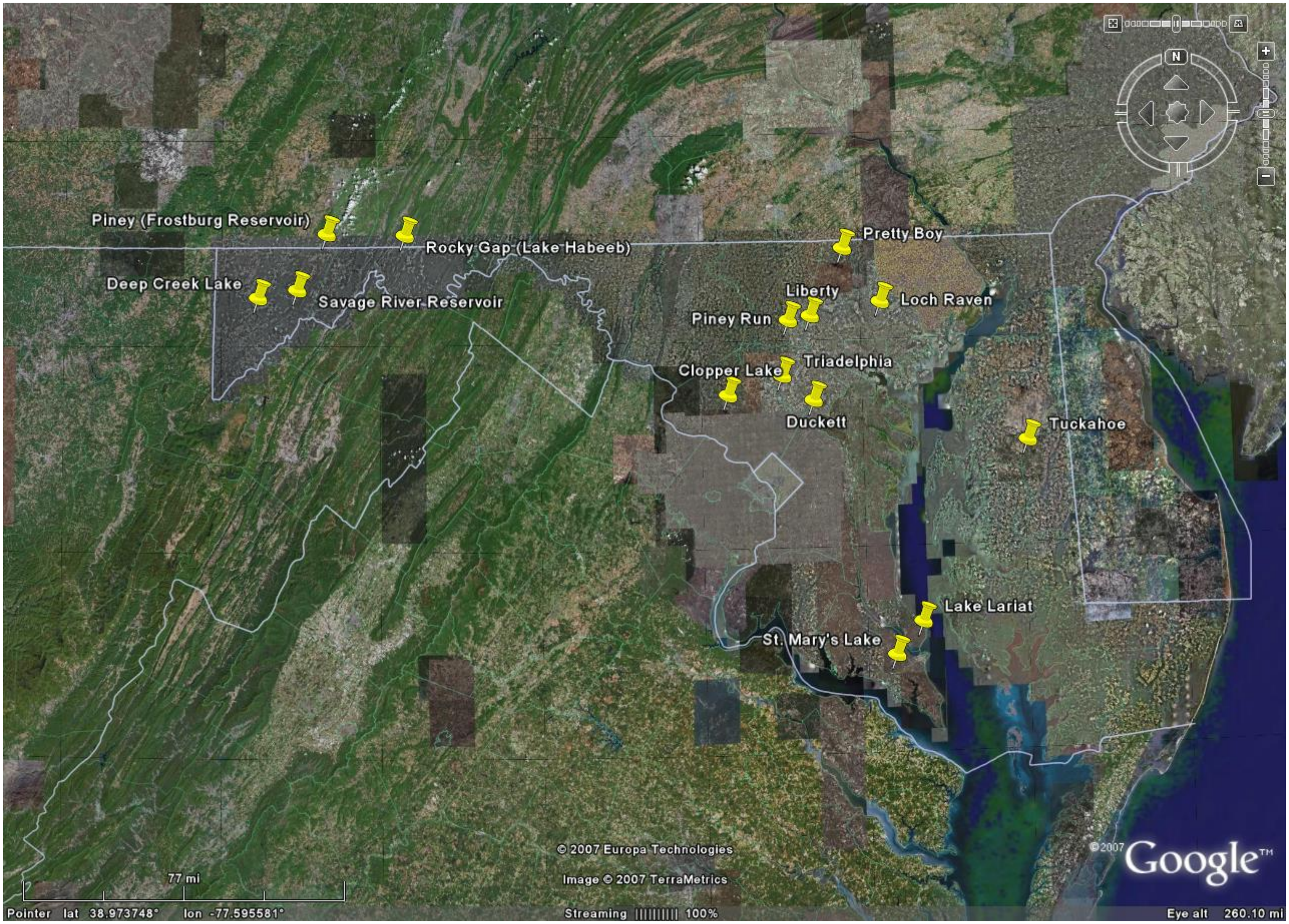


Figure 1. Location of the 14 study reservoirs in Maryland.

Table 1. Maryland reservoirs examined in this study.

Reservoir	Year Completed/ Modified	Normal Maximum Depth (m)	Surface Area (km2)	Normal Capacity (m3)	Watershed Area (km2)	Avg Annual Discharge (m3/sec)	Hydraulic Residence Time (y)	Watershed	County
Clopper	1975	12	0.36	2.00E+06	7	0.17	0.38	Seneca Creek	Montgomery
Deep Creek	1925	25	18.00	1.10E+08	163	8.66	0.40	Youghiogeny	Garrett
Duckett Res.	1953/1986	22.6	3.13	2.10E+07	343	2.38	0.28	Patuxent	Howard/Mont/PG
Lake Lariat	1965	9.1	0.39	1.90E+06	7	0.09	0.71	Patuxent	Calvert
Liberty	1953	44	12.57	1.60E+08	424	5.46	0.93	Patapsco	Carroll
Loch Raven	1923/86	23.2	9.71	9.00E+07	789	8.60	0.33	Gunpowder	Baltimore
Piney (Frostburg Res)	1934/1990	9.8	0.49	1.70E+06	31	0.65	0.08	Youghiogeny	Garrett
Piney Run Lake	1990	54.5	1.21	9.56E+06	28	0.36	0.84	Patapsco	Carroll
Pretty Boy	1936/1936	30	6.07	7.40E+07	206	2.90	0.81	Gunpowder	Baltimore
Rocky Gap (L. Habeeb)	1969/1988	25	0.85	6.60E+06	23	0.34	0.62	Evitt's Creek	Alleghany
Savage	1952	46.1	1.46	2.50E+07	270	4.17	0.19	N Branch Potomac	Garrett
St Mary's Lake	1975	6.4	1.01	3.90E+06	22	0.26	0.48	Lower Potomac	St. Mary's
Triadelphia Res.	1943/1999	15.8	3.24	2.30E+07	205	2.35	0.31	Patuxent	Howard/Mont
Tuckahoe	1975	2.7	0.35	3.20E+04	223	2.78	3.65E-04	Tuckahoe	Caroline

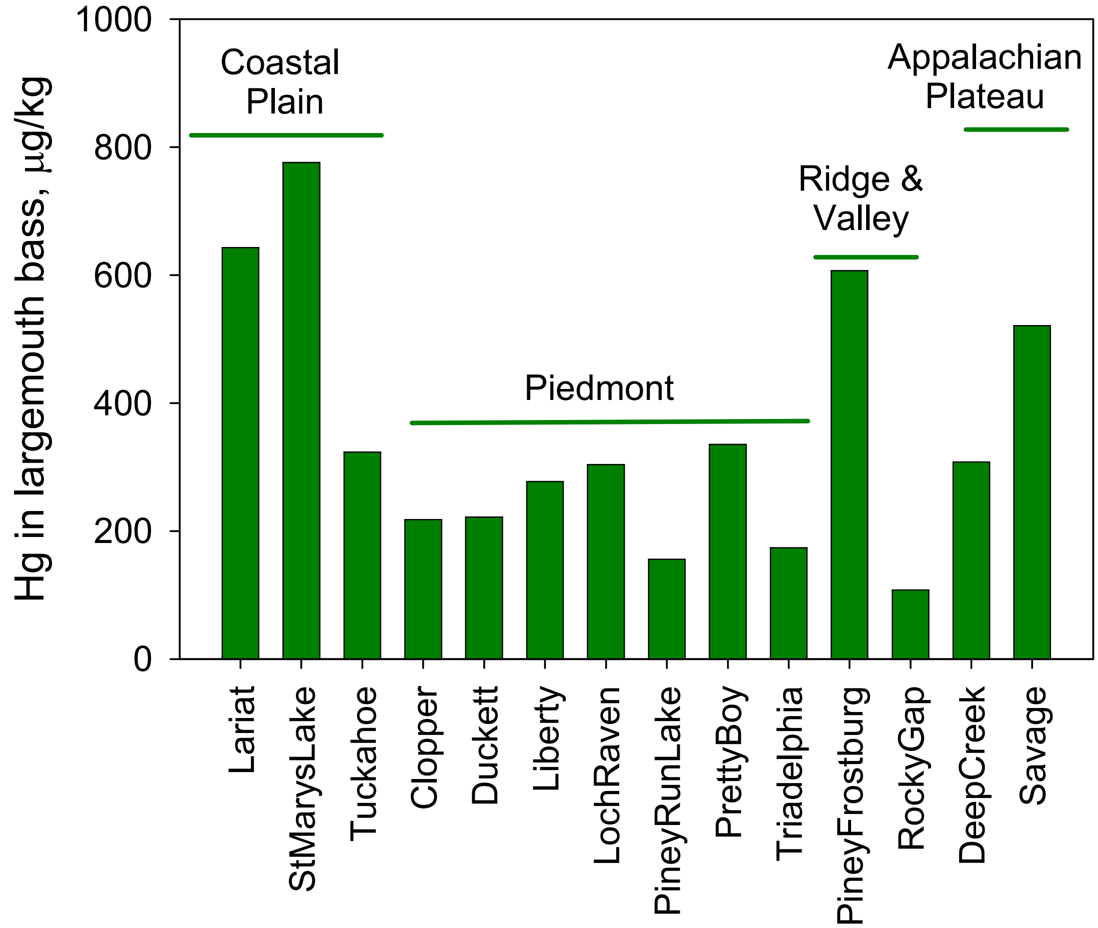


Figure 2. Mercury concentrations in largemouth bass in Maryland reservoirs. Data are size-normalized to a 370 mm fish (data from Sveinsdottir and Mason, 2005).

Table 2. Physiographic provinces for reservoirs in the study.

Reservoir	Physiographic Province	Subdivision
Clopper	Piedmont Plateau	Upland Section
Deep Creek	Appalachian Plateaus	
Duckett Res.	Piedmont Plateau	Upland Section
Lake Lariat	Coastal Plain	Western Shore Uplands
Liberty	Piedmont Plateau	Upland Section
Loch Raven	Piedmont Plateau	Upland Section
Piney (Frostburg Res)	Ridge and Valley	
Piney Run Lake	Piedmont Plateau	Upland Section
Pretty Boy	Piedmont Plateau	Upland Section
Rocky Gap (Habeeb)	Ridge and Valley	
Savage	Appalachian Plateaus	
St Mary's Lake	Coastal Plain	Western Shore Uplands
Triadelphia Res.	Piedmont Plateau	Upland Section
Tuckahoe	Coastal Plain	Delmarva Peninsula Region

Table 3. Land use for the watersheds of each of the study reservoirs. Land use data from MDE (TMDL reports), and from VERSAR.

Reservoir	Developed	Agriculture	Forest	Wetland
Clopper	77	1	17	1
DeepCreek	20	20	48	5
Duckett	2	56	37	3
Lariat	73	3	19	4
Liberty	25	41	31	1
LochRaven	19	42	37	1
PineyFrostburg	5	37	57	0
PineyRunLake	24	50	22	2
PrettyBoy	13	48	34	0
RockyGap	9	7	80	0
Savage	2	15	82	1
StMarysLake	8	8	79	5
Triadelphia	1	63	32	3
Tuckahoe	4	61	19	15

Background

Fish consumption advisories were issued for all freshwater lakes in the state by Maryland Department of the Environment (MDE) in December 2001. Human, wildlife and regulatory concerns prompted this study into the factors that contribute most to the elevated levels of MeHg in fish in many Maryland reservoirs. Some of these factors may be controllable through emissions, reservoir or land use management.

An ecosystem's sensitivity to Hg loading can be defined as the ability of that ecosystem to transform inorganic Hg load into MeHg in biota, as outlined in the conceptual diagram shown in Figure 3. Therefore, the bioaccumulation of MeHg depends on:

- Mercury loading rates
- Mercury transport to zones of methylation
- Controls on net methylation, and
- Controls on MeHg bioaccumulation

Many of the factors that impact Hg bioaccumulation are understood, although it has been difficult to create models that incorporate multiple factors, or that can be used to predict responses of individual aquatic systems. Many of these factors are quite variable among ecosystems. Some are more straightforward, particularly the loading of Hg to a system. In the sections below, we review the key processes known to control Hg bioaccumulation.

Mercury loading. Atmospheric Hg loading is the dominant Hg input to most, if not all, Maryland lakes (MDE TMDL studies; Mason et al., 1997a; 1997b; 1999; Mason et al., 2000b). Maryland watersheds are downwind from significant local and regional sources of Hg that is emitted from combustion sources. In the late 1990s and the early part of this decade, data from a limited number of short-term wet deposition monitoring sites in Maryland suggested that Hg deposition rates are relatively high statewide, but that there are significant spatial differences in deposition rate (Mason et al., 1997a; Mason et al., 2000b). Mercury Deposition Network (MDN) sites were set up in Frostburg and Beltsville, MD in mid-2004, and at the Smithsonian Environmental Research Center in late 2006. The first year of record (June 2004-June 2005) shows nearly double the amount of Hg deposition at the Beltsville site (12.0 ug/m2 y) than in Frostburg (6.7 ug/m2 y; Figure 4; Table 4). Both Hg concentrations in precipitation and total wet deposition rates were higher at Beltsville. This is consistent with measurements made during 1997-1998 by Mason et al. (2000) showing deposition rates at a site near Baltimore roughly double those at sites in southern Maryland and the Eastern Shore. Table 4 compares MDN data for all active sites in MD, VA and PA. Deposition rates range over about a factor of two. However, all sites other than Beltsville are situated outside urban areas, and away from near-field sources.

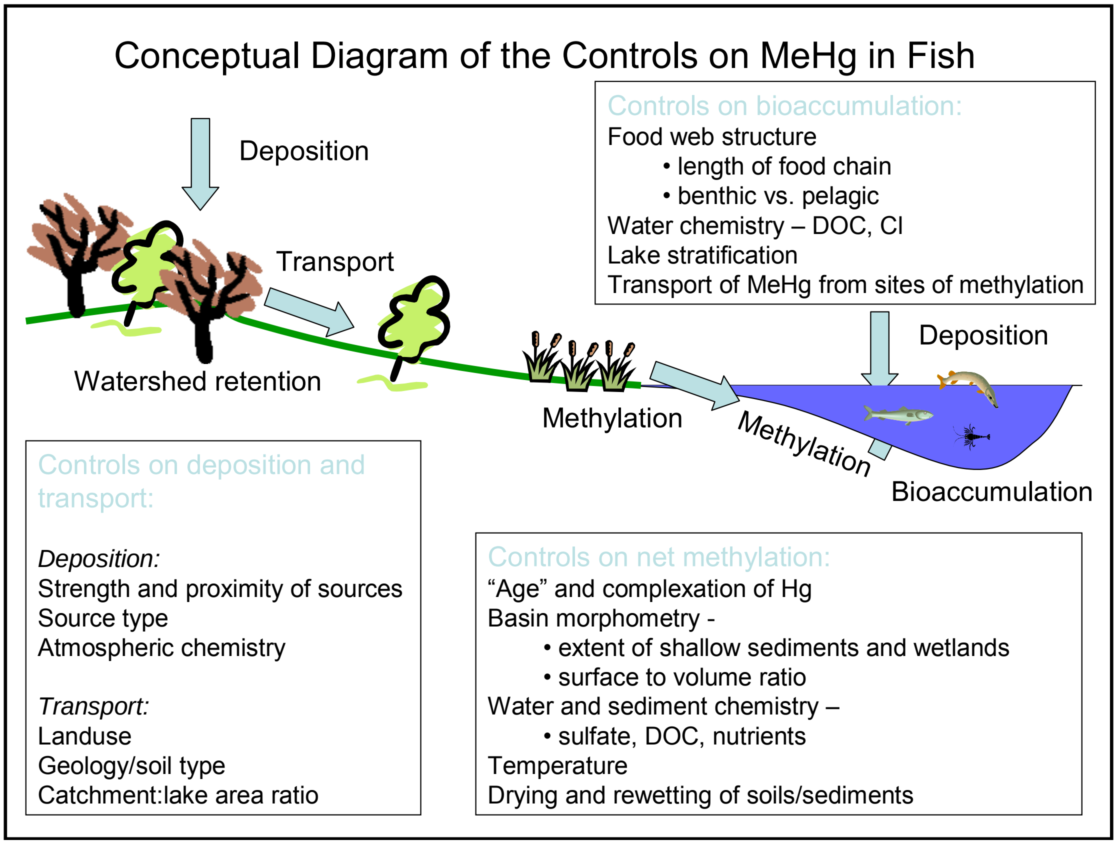


Figure 3. Conceptual diagram of the component processes that affect mercury accumulation in biota.

Figure 4. Data from Mercury Deposition Network sites in Maryland for 2004-2005. MD08 is in Frostburg, MD99 is in Beltsville. Top, precipitation data; middle, areal Hg deposition data; bottom, total Hg concentration in wet deposition.

Table 4. Annual Hg deposition for MDN sites in MD, VA and southern PA, June 2004-June 2005. The time period was chosen to match available period of record for new MDN sites in MD.

Site	Location	Hg deposition, µg/m2
MD08	Frostburg, MD	6.7
MD99	Beltsville, MD	12.0
VA08	Culpepper, VA	6.9
VA28	Shenandoah NP, VA	8.2
VA98	Gloucester Co, VA	New site
PA00	Adams Co, PA	8.2
PA37	Holbrook, PA	9.2
PA47	Millersville, PA	7.5
PA60	Valley Forge, PA	10.2

Mark Garrison of VERSAR, through the Maryland Power Plant Research Program has recently applied the CALPUFF modeling system to estimate wet and dry Hg deposition rates across Maryland (Sherwell et al. 2006). This model was based on an inventory of local sources (including power plants - Figure 5 - and other sources), and developed atmospheric chemistry modules for the model that resulted in significant dry and wet deposition. Details of model construction can be found in Sherwell et al. 2006. The model shows large gradients in deposition rates across Maryland and significant hot spots, especially around Baltimore (Figures 6 -8). Modeled total Hg deposition for each of the reservoirs is shown in Table 5.

Attempts to relate Hg concentrations in fish to deposition patterns have historically been difficult (Wren and MacCrimmon, 1983; Wiener et al., 1990; Hakanson et al., 1998; Cope and Wiener, 1990), in part because of the many other factors that affect Hg transport, methylation and bioaccumulation in watersheds. Recently, however, a small number of studies have successfully done so in the US and Europe, because of increased data availability from wet deposition and fish monitoring programs.

One of the best historical spatial examples is the south to north gradient in Hg in fish in Sweden, driven by patterns of Hg emissions from eastern Europe (e.g., Lindqvist et al. 1991, Meili et al. 2003; Munthe et al. 2004). However, specific ecosystem characteristics also contribute to variability in fish Hg between regions. This variation is particularly evident for the southernmost part of Sweden, where very low concentrations of Hg were found in fish despite a larger influence of atmospheric pollution. This region of Sweden has mainly agricultural land use. This land disturbance typically increases lake productivity, decreasing fish Hg concentrations through biodilution (Pickhardt et al. 2002).

Recently, a temporal pattern has also emerged in northern Europe, following the decline of older, industrial facilities without environmental controls in former Soviet Union countries. Johansson et al. (2001) reported a 20% average decline in Hg concentrations for northern pike (*Esox lucius*, 1 kg standard) from 42 remote Swedish lakes sampled initially between 1981-87, and again between 1988-95. Wet Hg deposition declined by more than 50% in southern Sweden (less in central and northern parts) during this period. Although fish Hg concentrations declined on average for the study lakes, increases and decreases were both observed for fish Hg levels in individual water bodies. Other observations suggest that multiple factors likely affect fish Hg concentrations (Munthe et al. 2007) and demonstrate the need for carefully designed monitoring programs that monitor not just Hg, but also sufficient environmental information to help explain very different trends that might emerge among individual sites (Harris et al. 2007).

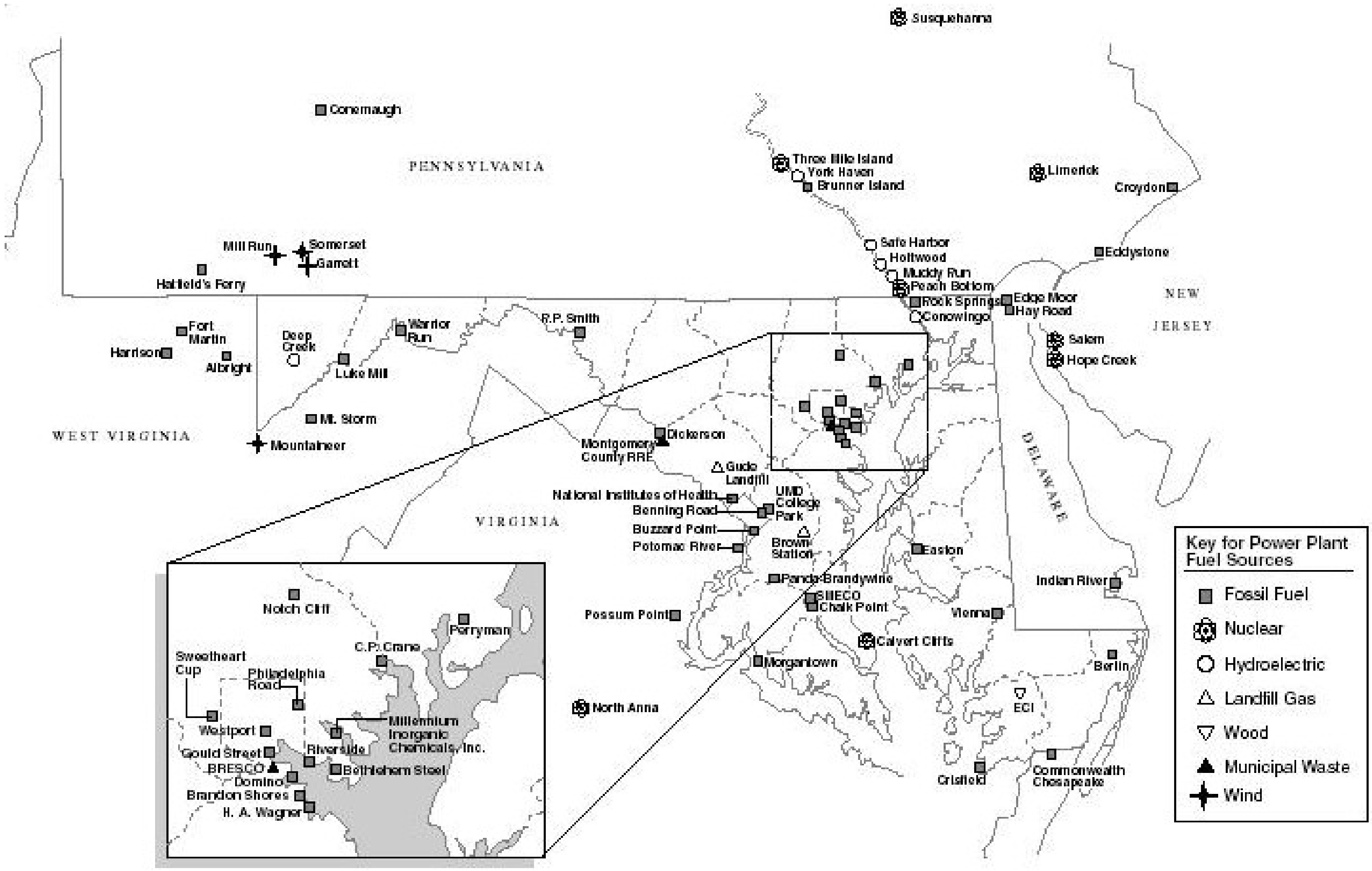


Figure 5. Location of electric generation plants in Maryland. Source: Maryland DNR, MD Power Plant Research Program. http://esm.versar.com/pprp/factbook/plantlocations.htm

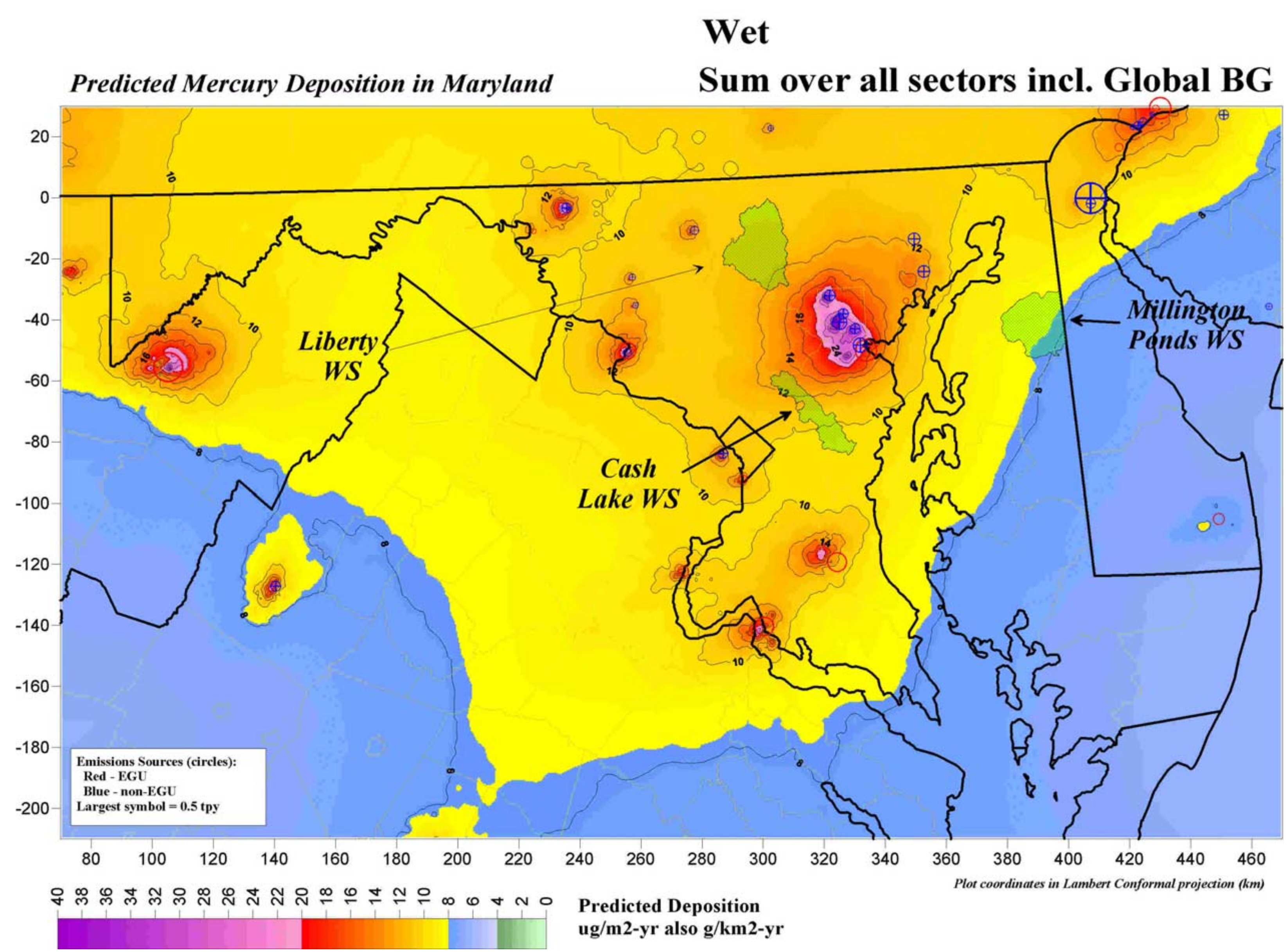


Figure 6. Modeled wet mercury deposition rates across Maryland (from Sherwell et al. 2006).

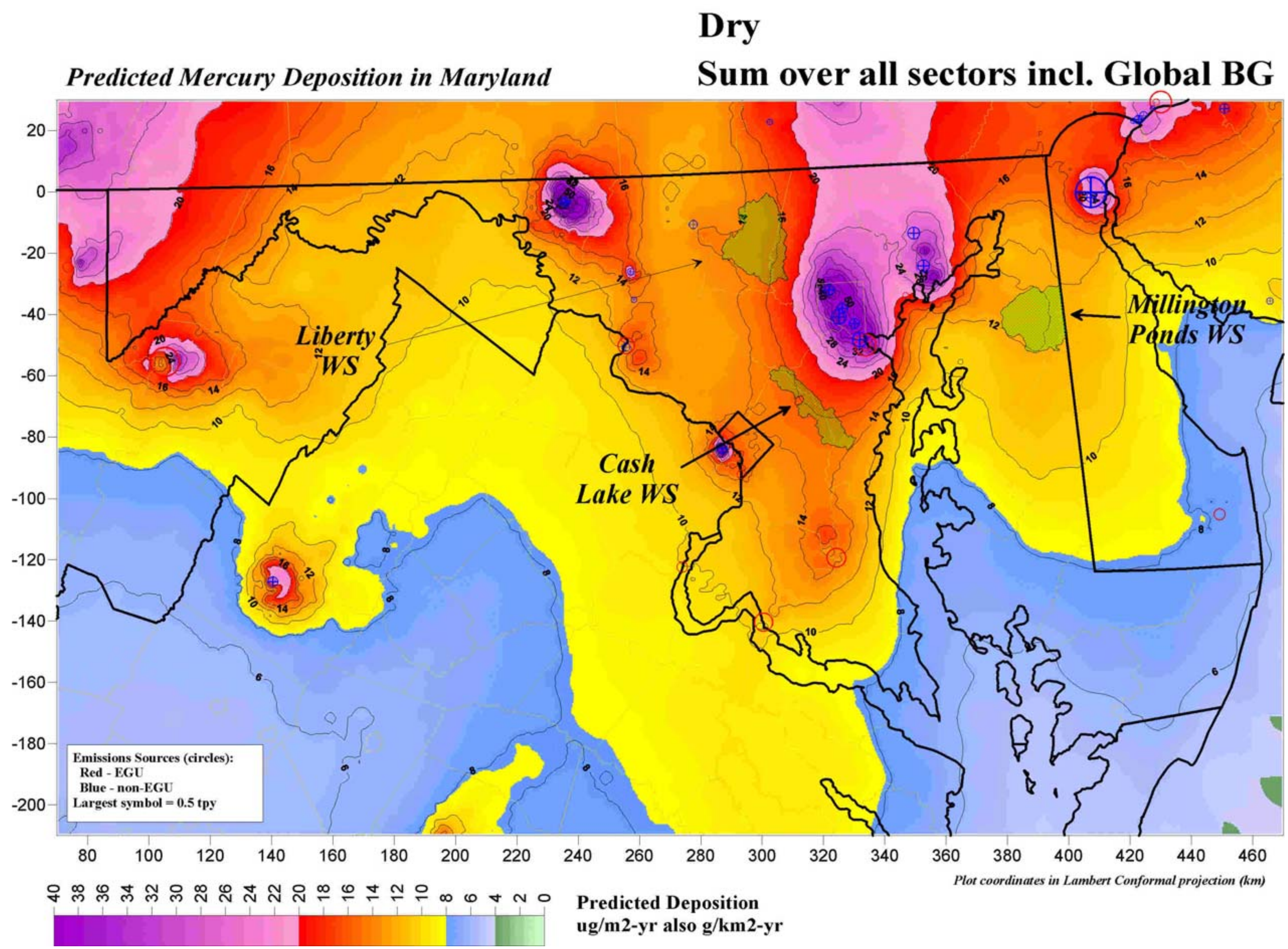


Figure 7. Modeled dry mercury deposition rates across Maryland (from Sherwell et al. 2006).

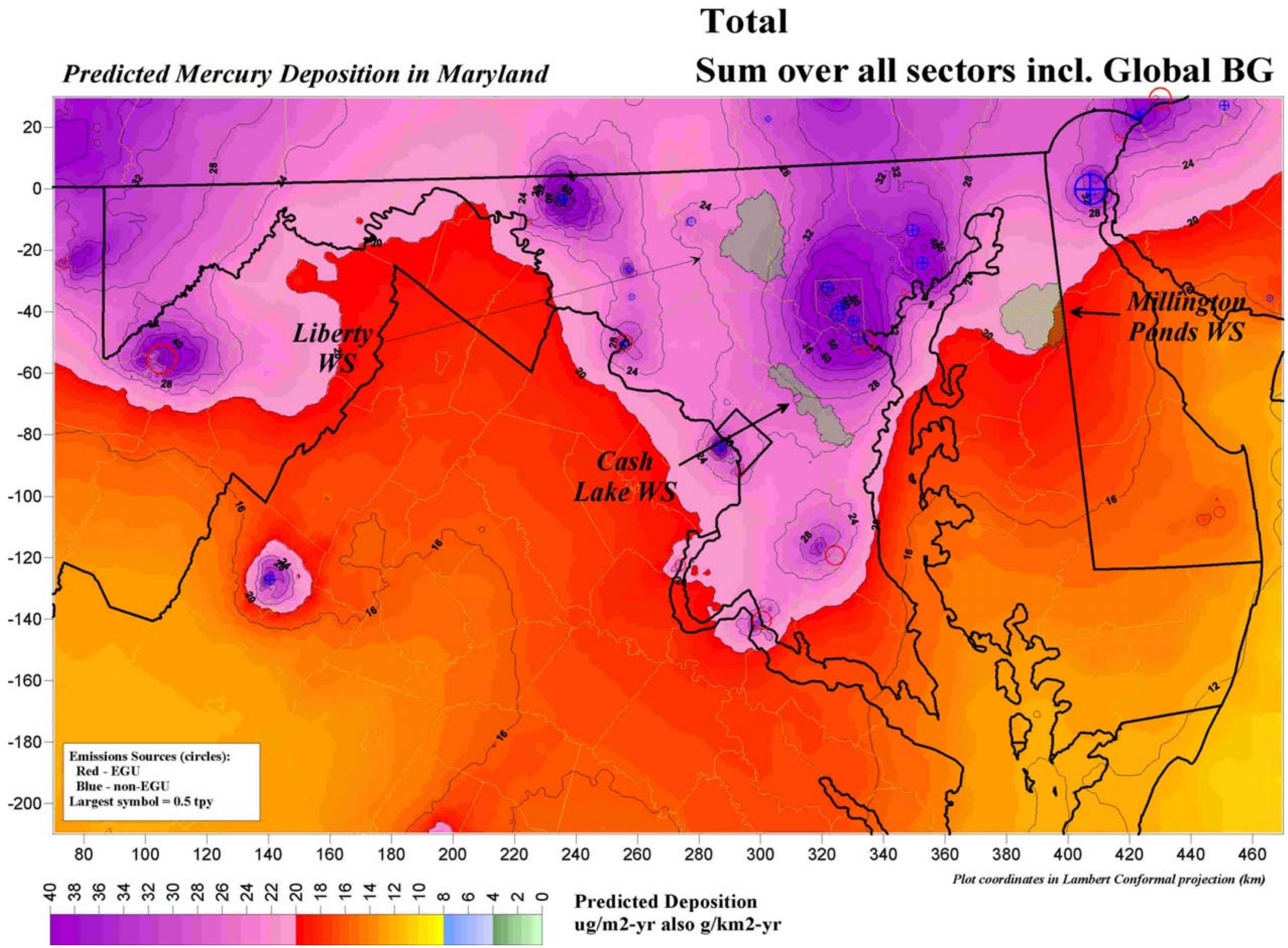


Figure 8. Modeled total mercury deposition rates across Maryland (from Sherwell et al. 2006).

Table 5. Modeled total Hg deposition rates for Maryland reservoirs, estimated from Sherwell et al. 2006.

Reservoir	ug/m2 y
Clopper	24
DeepCreek	29
Duckett	27
Lariat	18
Liberty	28
LochRaven	37
PineyFrostburg	27
PineyRunLake	26
PrettyBoy	33
RockyGap	22
Savage	29
StMarysLake	18
Triadelphia	28
Tuckahoe	20

In the US, a number of large spatial datasets including atmospheric deposition, surface waters, sediments and biota were recently compiled for eastern North America as part of a Northeast Ecosystem Research Cooperative (NERC) initiative (Evers and Clair 2005a, 2005b). None of the organisms studied showed direct relationships with spatial patterns in atmospheric Hg deposition, except blood Hg of Bicknell's thrush for which Rimmer et al. (2005) found a strong relationship with estimates of Hg in litterfall.

However, biological Hg hotspots were geographically identified within these datasets (Evers et al. 2007) . Yellow perch and common loon were chosen as indicator species for human and ecological effects of Hg, respectively. Impact thresholds of 0.30 µg/g (wet weight) for yellow perch fillets and 3.0 µg/g (wet weight) in blood for common loons were used to determine the location of biological Hg hotspots. These biological Hg hotspots reflect conditions that influence ecosystem and associated biological response to atmospheric Hg deposition. In particular three factors were identified that control the formation of biological Hg hotspots, including local elevated atmospheric Hg deposition due to proximity to emission sources, landscape sensitivity (related to forest cover, shallow hydrologic flowpaths, the abundance of wetlands and unproductive surface waters), and water level manipulations. A case study in southeastern New Hampshire demonstrates that local Hg emissions contribute significantly to local deposition. Historic data and model projections for southern New Hampshire showed that Hg emission reductions resulted in decreased Hg deposition and rapid recovery from Hg in aquatic biota (Evers et al. 2007).

Recent studies by Hammerschmidt and Fitzgerald (2005; 2006) reported strong relationships between rates of wet deposition of Hg and Hg in both mosquitoes and in largemouth bass across North America. Mercury in mosquitoes is relevant to aquatic ecosystems because these animals have aquatic life stages. Since more than 90% of the Hg in mosquitoes occurred as MeHg, this data set also provides a link between inorganic Hg deposition and methylation occurring in the aquatic ecosystem. Fish data from the US EPAs data set as well as individual state monitoring programs were used in the analysis. Both of these studies made use of the full U.S. Mercury Deposition Network data set for the first time in this context.

Mercury in Watersheds. The biophysical characteristics of watersheds control the retention of Hg in the terrestrial compartment and transport of Hg to sites of methylation via complex hydrologic, chemical, and biologic processes. However, evidence exists suggesting that there are generalizable watershed characteristics that integrate the key processes affecting the efficiency with which Hg is transported through the landscape to zones of methylation, and that these characteristics can be readily measured. Some of these key characteristics are: the size and topography of the watershed (affecting the residence time and flow pathways of runoff); land cover (affecting dry deposition rates, and the degree of interaction between water and both methylating and non-methylating soils); and land-use, governing the relative importance of particulate Hg load to sites of methylation (e.g. Balogh et al. 2005; Hurley et al. 1995; Warner et al. 2005).

Watershed size is an important determinant of Hg retention and delivery to aquatic ecosystems. Based on a synthesis of data from watersheds of varying size, Grigal (2002) suggested that there is an overall decrease in Hg flux and concentrations in runoff with increasing watershed size due to less efficient transport and increased loss processes. Very large watersheds will thus be much less responsive to changes in atmospheric Hg load than those where the watershed influence is smaller. Watersheds subject to significant transport of sediment in surface runoff such as agriculturally-dominated systems contribute larger amounts of Hg to sites of methylation (Warner et al. 2005) than forested watersheds (Hurley et al.1995; Babiarz et al. 1998). However, as this Hg is largely associated with particulate matter, it may be less bioavailable than dissolved inputs of Hg.

When Hg loading is affected by land use, particularly in landscapes with erodable soils (Balogh et al. 2005), land use practices can have a direct effect on MeHg in biota. Using a modeling approach, Roué-Le Gall et al. (2005) showed that when watershed characteristics were coupled to information about the food web in 45 lakes, the sensitivity of biota to MeHg contamination could be predicted qualitatively (high to low).

The common factor among all of the watershed characteristics identified above is soil cover. Soils retain Hg in watersheds, and this retention is strongly coupled to the organic matter fraction (Grigal, 2002) where Hg is stored either through sorption of Hg deposited directly from the atmosphere, or associated with organic matter derived from the forest canopy or floor. Even in landscapes characterized by thin and discontinuous soils, newly deposited Hg appears to be nearly completely retained in the short term (Harris et al. 2007). The magnitude and timing of the release of Hg from this pool is controlled by the rate of decomposition of the soil organic matter pool, and physical removal of the soil itself through erosion. The current lack of insight in the dynamics of Hg release from soils makes the greatest contribution to the uncertainty in the quantitative prediction of the magnitude and timing of the effects of a change in Hg load on MeHg in biota.

Hg methylation. In order for Hg loads to ecosystems to result in MeHg in biota, inorganic Hg must be converted into MeHg. Net MeHg production is affected by a complex system of controls, most importantly:

- the areal extent and connectivity of methylating zones within the ecosystem;
- the bioavailability of Hg delivered to those zones for uptake and methylation by micro-organisms; and
- the type and activity of methylating and demethylating bacteria within zones of net methylation.

The landscape compartments that support MeHg production in watersheds are reasonably well defined. Because methylation takes place under anoxic conditions (Benoit et al. 2003), the areal extent of wetlands and hydric soils (St. Louis et al. 1994; Hurley et al. 1995; Babiarz et al. 1998) is a strong determinant of MeHg export from watersheds. Wetlands can be particularly active zones of MeHg production (e.g. the Florida Everglades, Gilmour et al. 1998; tidal mashes, Marvin-DiPasquale et al. 2003). Methylation in bogs (Benoit et al. 1998), fens (Branfireun et al. 1996; Heyes et al. 2000) and riparian zones (Bishop et al. 1995; Driscoll et al. 1998; Krabbenhoft et al. 1995) is highly dependent on flow paths and hydrologic connectivity that govern the location and extent of zones of microbial activity. Forestry operations have been shown to increase the load of MeHg to the aquatic ecosystem (Garcia and Carignan, 1999; Porvari et al., 2003; Munthe and Hultberg, 2004). In freshwater aquatic ecosystems, shallow, organic-rich lake sediments are often major zones of methylation (e.g. Krabbenhoft et al. 1998; Kainz et al. 2003), and therefore lake surface to volume ratio impacts the conversion of Hg to MeHg (Bodaly et al. 1993). The presence of anoxic bottom waters in stratified lakes significantly enhances MeHg production (Watras et al. 1995; Eckley et al. 2005).

Flooding (Lucotte et al., 1999, Heyes et al. 2000; St. Louis et al. 2005) and soil drying and rewetting cycles (Krabbenhoft 2001; Rumbold et al. 2006) strongly impact methylation, in large part through the sulfur cycle. Oxidation of reduced sulfur during drying leads to a pulse of sulfate reduction and Hg methylation when soils rewet (Gilmour et al. 2004). Changes in water levels in lakes and wetlands can significantly impact MeHg levels in fish (i.e., Sorensen et al. 2005). Several examples of this behavior from reservoirs are also available (Verta et al. 1986, Snodgrass et al. 2000, Haines and Smith 1998, Evers et al. 2004).

A few key biogeochemical cycles have a large impact on MeHg production. In many cases, however, the relative impact of these cycles on the activity of methylating microorganisms vs. the bioavailability of Hg to these cells is poorly understood.

- *Sulfur.* The Hg and S cycles are intimately linked, thus linking acid rain to the Hg cycle. The balance between sulfate and sulfide is a key control on Hg methylation rate in many ecosystems. Sulfate stimulates Hg-methylating sulfate reducing bacteria (SRB), while excess sulfide creates mercury complexes that are not bioavailable. Sulfate-stimulation of methylation has been demonstrated in studies that range from pure culture (King et al. 2000; Benoit et al. 1999), to sediment and soil amendments (Compeau and Bartha 1985; Gilmour et al. 1992; Harmon et al. 2004; King et al. 2001; Benoit et al. 2003), to field amendments to lakes and wetlands (Watras et al. 1994; Branfireun et al. 1999; Benoit et al. 2003). Among these studies, the optimal concentration for methylation ranges from 10 to about 300 uM sulfate, while the optimal sulfide concentration is quite low, about 10 uM. Factors like iron and organic matter concentration that impact Hg and S complexation change these optima.

- *pH.* Many studies have linked lake acidity to increased MeHg bioaccumulation (e.g., Grieb et al. 1990; Kamman et al. 2004). This observation has important implications for management of fish Hg contamination. Several mechanisms have been hypothesized, and pH may co-vary with Hg and sulfate loadings. This pattern could be driven by pH effects on bioaccumulation *per se*, for example decreases in aquatic productivity with decreases in pH. However, the effect could also be linked to MeHg production. Acidity linked to sulfate deposition may stimulate the activity of Hg-methylating SRB. Additionally, increased uptake of Hg by micro-organisms that use facilitated transport for Hg uptake increases with decreasing pH (Kelly et al. 2003).

- *Dissolved organic matter (DOM).* Both the character and concentration of DOM affect the complexation and potential bioavailability of Hg for methylation (Haitzer et al. 2002; Aiken et al. 2003; Miller 2006). Higher molecular weight DOM limited Hg availability to one Hg bioreporter (Barkay et al. 1997), probably through the formation of complexes that are too large to assimilate; while small organic ligands enhanced Hg uptake by another bioreporter through facilitated transport (Golding et al. 2002). Recent work suggests that DOM is an important ligand under sulphidic conditions, through interactions with HgS complexes (e.g. Ravichandran et al. 1998; Miller 2006). The relationships between DOM and methylation need to be further explored before they can be adequately modeled.

- *Iron.* Like S and DOM, the impact of Fe on methylation appears to be concentration and environment dependent. (Warner et al. 2003, 2005; Mehrota et al. 2005). Impacts on net methylation may occur via Hg complexation or microbial activity. A few strains of Fe(III)-reducing bacteria are now known to be capable of Hg methylation (Fleming et al. 2006), but the impact in the environment needs further study.

- *Hg "aging."* Recent Hg-amendment studies in lakes and wetlands show that Hg bioavailability for methylation decreases as Hg "ages" in sediments and soils (Orihel et al. 2006; Harris et al. 2007). Understanding the rate of ageing will be key to modeling ecosystem responses to changes in Hg load.

- *Type and activity of bacteria.* Although Hg methylation can be measured in almost any soil or sediment under reducing conditions, only a few sulfate-reducing bacteria (see Benoit et al. 2003), and a few closely-related Fe-reducing bacteria (Fleming et al. 2006) have demonstrated methylation ability in pure culture. There are a variety of microbial demethylation mechanisms, including the *mer* operon detoxification system that is spread widely among micro-organisms in contaminated environments, and may serve to limit MeHg accumulation at high Hg concentrations. The oxidative demethylation pathway (Oremland et al. 1991) is linked to one-carbon metabolic pathways, and is likely tied to overall carbon utilization rates rather than to Hg levels. When observed together through time or space, demethylation rates vary substantially less than methylation rates, and thus MeHg concentrations are often well-correlated with methylation rates alone (Marvin-DiPasquale et al. 1998; Benoit et al. 2003).

Bioaccumulation. Once formed, there are important physical, biological and chemical controls on MeHg bioaccumulation that dramatically impact the transfer of Hg load into MeHg in fish and other predators. Differences in these processes between different lakes may result in largely different responses to Hg loading.

Biophysical controls. Methylmercury uptake is impacted by the nature (pelagic vs. benthic) and structure of the food web relative the location of MeHg production (e.g. Gorski et al. 2003). Marshes like the Everglades provide a good example of benthic-driven ecosystems, and in which the lower portion of the food web is in direct contact with the major zone of methylation in soils (Cleckner et al. 1998; Gilmour et al. 1998). Lakes with MeHg production in anoxic bottom waters (Watras et al. 1995, Eckley et al. 2005) illustrate the transfer of MeHg to pelagic aquatic food webs, often providing a pulse of MeHg to surface waters and plankton after fall turnover (e.g. Herrin et al. 1998). In large lakes and oceans, MeHg accumulation occurs predominantly around coastal areas of MeHg production (e.g. Manoloupolos et al. 2003). Zones of MeHg production that are disconnected from aquatic food webs (e.g., isolated bogs) may have little impact on Hg in aquatic biota, but can impact terrestrial food webs (Banks et al. 2005; Evers et al. 2005).

Biological controls. Food web structure, fish population age structure, and physiological controls on uptake all impact the bioaccumulation of MeHg. Any examination of change in MeHg in fish through time must examine potential concomitant changes in food web and fish population structure. Changes in Hg bioaccumulation patterns following the invasion of a fish species provide an example of how food web structure impacts Hg bioaccumulation (Swanson et al. 2003). Food web structure and composition also impact bioaccumulation through food quality (i.e., Lawson and Mason 1998, Lucotte et al. 1999) and gut chemistry (e.g., Laporte et al. 2002) effects on uptake efficiency.

Chemical controls. The bioavailability of MeHg to organisms at the bottom of food webs is affected by MeHg complexation (Wiener et al. 1990a,b., Scheuhammer et al. 2007). The inhibition of MeHg accumulation by higher molecular weight DOM has been well documented for fish (e.g. Lang et al. 1993) and zooplankton (e.g. Back et al. 1995). The role of DOM overall in Hg transport, methylation and bioaccumulation is complex, but central to the physical and biogeochemical behavior of Hg in watersheds. For example, complexation with small organic molecules can enhance uptake of MeHg (Lawson and Mason 1998). An inverse relationship between pH and MeHg bioaccumulation is also well documented - for example for loons (Meyer et al. 1998), benthic invertebrates (Rennie et al. 2005) and fish (e.g. Roué-Le Gall et al. 2005; Chen et al. 2005).

Study Design and Methods

Objective: To identify the key parameters that affect methylmercury (MeHg) production and bioaccumulation in MD reservoirs.

Overall Design. We sought to construct a data set for Maryland reservoirs that would be sufficient to test hypotheses about the influence of key variables on MeHg production and bioaccumulation in Maryland lentic freshwaters. Some of the needed information was available prior to the study, particularly fish Hg concentrations for many Maryland reservoirs from two studies (Gilmour and Riedel 1999; Svensdottir and Mason 2005) that gave very comparable information. Substantial new information was collected, particularly water and sediment chemistry for all of the study lakes. Additionally, physical information about the reservoirs and watersheds was compiled from other sources.

Once compiled, statistical approaches were used to examine the data set. Size-normalized largemouth bass Hg concentrations were used as the ultimate dependent variable. However, other variables were used to examine specific components of the biogeochemical Hg cycle. For example, Hg concentrations in water and sediment were used as indicators of Hg deposition and transport; the ratio MeHg/Hg in water and sediment was used as an indicator of methylation efficiency; and the ratio of MeHg in fish to MeHg in water was used to assess MeHg bioaccumulation. Each of these variables was regressed against the suite of variables that are likely to affect each process, using single and multiple regressions.

Database Parameters. The parameters chosen for inclusion in the database and source of these data are given in Table 6. These parameters were chosen based on the conceptual diagram shown in Figure 2 and on the discussion in the background section above.

Study Sites. The fourteen Maryland reservoirs examined in this study are listed in Table 1, along with some of their hydrological and morphometric characteristics. All freshwater bodies of any size in Maryland are man-made impoundments. The chosen reservoirs spanned a wide range of key parameters, including land use (Table 3) and location within Maryland. Study reservoirs were chosen in of all Maryland's physiographic provinces except the Blue Ridge (Table 2). The final choice of lakes for the database was based on the availability of size-normalized largemouth bass data.

Table 6. Parameters used to examine the controls on MeHg bioaccumulation in MD reservoirs, and the source of those data. N/A = not available.

Database Parameters	Data source
Factors that influence Hg Deposition and Transport:	
Hg deposition rate	MDE VERSAR CALPUFF model
Watershed size	MDE, VERSAR
Watershed: waterbody surface area ratio	MDE, VERSAR
Reservoir capacity	MDE TMDL reports, WSSC
Hydraulic residence time	Flow data from USGS, WSSC, MDE
Physiographic province	MGS
Land use, overall and buffer areas	MDE, VERSAR
Reservoir water level fluctuation	Data not compiled
Factors that influence net MeHg production:	
temperature	
Basin morphometry -	
Lake size	MDE, WSSC, VERSAR
Lake stratification and anoxia	This study
Water chemistry -	
Hg concentration	This study; Svensdottir and Mason 2005
pH, sulfate, DOC, conductivity, TSS	This study; Svensdottir and Mason 2005, MDE
trophic status (nutrients)	This study; Svensdottir and Mason 2005, MDE
Sediment chemistry -	
Hg concentration	This study
Sediment:water partitioning of Hg	This study
Pore waters: pH, sulfate, sulfide, Fe, Mn, DOC	This study
bulk phase: density, organic matter, reduced sulfur (AVS/CRS)	This study
Factors that influence MeHg bioaccumulation:	
Food web structure-	
Length of food web	Data not available
Benthic vs pelagic-based food web	Data not available
Water chemistry -	
MeHg concentration	This study; Svensdottir and Mason 2005
pH, DOC, TSS	This study; Svensdottir and Mason 2005
trophic status (nutrients)	This study; Svensdottir and Mason 2005, MDE
Dependent variables:	
Size-normalized Hg in largemouth bass	Svensdottir and Mason 2005; Gilmour & Riedel 1999
Hg and MeHg in surface waters	This study; Svensdottir and Mason 2005
Hg and MeHg in sediments and sediment pore waters	This study

Sampling and Analysis Methods. Sampling and analysis methods for the water and sediment sampling conducted during 2003/2004 for the 10 reservoirs sampled by SERC/CBL are discussed in this section. Methodologies for the reservoirs sampled by University of Maryland Appalachian Laboratory (Savage, Deep Creek Lake, Piney Frostburg and Lake Habeeb) are described in Castro (2006).

Each reservoir was visited once during summer 2003 or 2004. For each reservoir, sediment samples were collected at three sites. Sediment samples were generally taken from three different water depths, 0.5-2 m, 3-5m, and the deepest point in the reservoir, with exceptions in shallow systems like Tuckahoe. Samples were collected using a 20 cm square Eckman dredge, deployed by hand from a small boat, which allowed collection of a minimally disturbed surface layer. The Eckman box cores were sub-sampled on the boat using 4.8 cm diameter PVC tubes. The top 4 cm of cores were subsequently sectioned for analysis. This approach was chosen, rather than sediment depth profile data, to allow the highest sample number for comparison among sites within the study budget.

Sediment cores were returned to the laboratory on ice within hours of collection and processed that day. The parameters measured in sediments are listed in Table 7, along with the analytical methods used. Sediment pore waters were separated and processed inside a N_2-filled glove bag with an air lock (Coy Laboratory Products) using H_2 gas and Pd-catalysts to maintain O_2-free conditions. Because most sediments are anoxic within mm of the surface, sediment processing under strict anaerobic conditions preserves redox-sensitive components like sulfide and Fe(II); and also preserves the redox sensitive partitioning of Hg and MeHg between solid and dissolved phases. Pore waters were separated by vacuum filtration, using acid-washed 0.2 um disposable CN/PC Nalge filter units. Trace-metal clean procedures were used throughout, using rigorously acid-cleaned sample containers and low-Hg acids where needed. All analyses were performed from composites of at least 3 sediment cores.

In each reservoir, water samples were collected at the deepest point in the reservoir (generally behind the dam) on the same day that sediments were collected. A depth profile of temperature, DO, conductivity and percent incident light was taken using YSI meters and dual LICOR light meters, *in situ*. Water samples for Hg, MeHg and other analytes were collected using Go-Flo bottles from two depths, one near surface and one near bottom. Sampling depths were chosen from the DO profile so that bottom samples were taken in anoxic bottom waters where present. The parameters measured in water are listed in Table 8, along with the analytical methods used.

Table 7. Sediment samples collected in eastern Maryland reservoirs, 2003/2004.

Bulk phase measurements	**Method**	**Analysis done by:**	**Reference**
Hg	HNO3/HCl digestion, isotope dilution ICP-MS	SERC	Gilmour et al. 1998; Hintelmann and Ogrinc 2003
MeHg	distillation, ethylation, isotope dilution ICP-MS	SERC	Gilmour et al. 1998; Hintelmann and Ogrinc 2003
Bulk density, dry weight, porosity, loss-on-ignition	Standard Methods	SERC	Standard Methods
Reduced sulfur complexes - AVS, CRS		SERC	Gilmour et al. 1998;
Extractable Fe(II)/Fe(III)	0.5 N HCl extraction, ferrozine, spectrophotometry	SERC	Roden and Wetzel 2002
Pore water measurements (all <0.2 um)			
pH	low ionic strength probe	SERC	Standard Methods
sulfide	preservation in fresh sulfide anti-oxidant buffer, ion-selective probe	SERC	Standard Methods
Hg	BrCl digestion, isotope dilution ICP-MS	SERC	Gilmour et al. 1998; Hintelmann and Ogrinc 2003
MeHg	distillation, ethylation, isotope dilution ICP-MS	SERC	Gilmour et al. 1998; Hintelmann and Ogrinc 2003
Fe, Mn	GFAA	SERC	Standard Methods
anions (F, Cl, NO3, PO4, SO4)	ion chromatography	SERC	Standard Methods
DOC	uv oxidation/IR CHECK	UMD/CBL analytical services	Standard Methods

Table 8. Water samples collected in eastern Maryland reservoirs, 2003/2004.

Filtered water:	**Method**	**Analysis done by:**	**Reference**
pH	pH probe	SERC	Standard Methods
sulfide	preservation in fresh sulfide anti-oxidant buffer, ion-selective probe	SERC	Standard Methods
Hg	BrCl digestion, ICP-MS	CBL	Hintelmann and Ogrinc 2003
MeHg	distillation, ethylation, ICP-MS	CBL	Hintelmann and Ogrinc 2003
NO3, NO2	Automated cadmium reduction	UMD/CBL analytical services	USEPA. 1979. Method No. 365
anions (Cl, SO4)	ion chromatography	SERC	Standard Methods
DOC	High temperature (680 degrees C) combustion	UMD/CBL analytical services	Sugimura, Y. and Y. Suzuki. 1988
Particulate:			
PC,PN	EPA method 440.0	UMD/CBL analytical services	USEPA. 1997. Method No. 440
Hg	HNO3/HCl digestion, ICP-MS	CBL	Heyes et al. 2004; Hintelman and Ogrinc 2003
MeHg	distillation, ethylation, ICP-MS	CBL	Heyes et al. 2004; Hintelman and Ogrinc 2003
TSS	filtration/gravimetric	CBL	Standard Methods
Chlorophylls	Fluorometric, 90% acetone extraction	UMD/CBL analytical services	Welschmeyer, N.A. 1994.

Results from reservoir sampling

This section presents the raw data from the 2003-2005 reservoir sampling, and highlights some of the key relationships among variables. Statistical analysis of the full 2000-2005 data set, including correlation coefficients for each variable pair, and stepwise linear regression models can be found in the next section.

Depth profiles of reservoirs sampled. Figures 9A-9J show depth profiles for the 10 eastern reservoirs on their sampling dates in 2003-2004. Summer depth profiles (where available) for the western Maryland reservoirs are shown in Figures 10 A-D. The water columns of all but two of the reservoirs sampled were stratified when sampled in the summer. Bottom waters were often anoxic, a condition that generally enhances MeHg production and bioaccumulation in lakes. The exceptions were Tuckahoe (on the eastern shore), which is a very shallow lake; and Savage Reservoir, which was tested in the fall when stratification would not be strongest. The western lakes tended to be less turbid, and fewer were fully anoxic at the bottom.

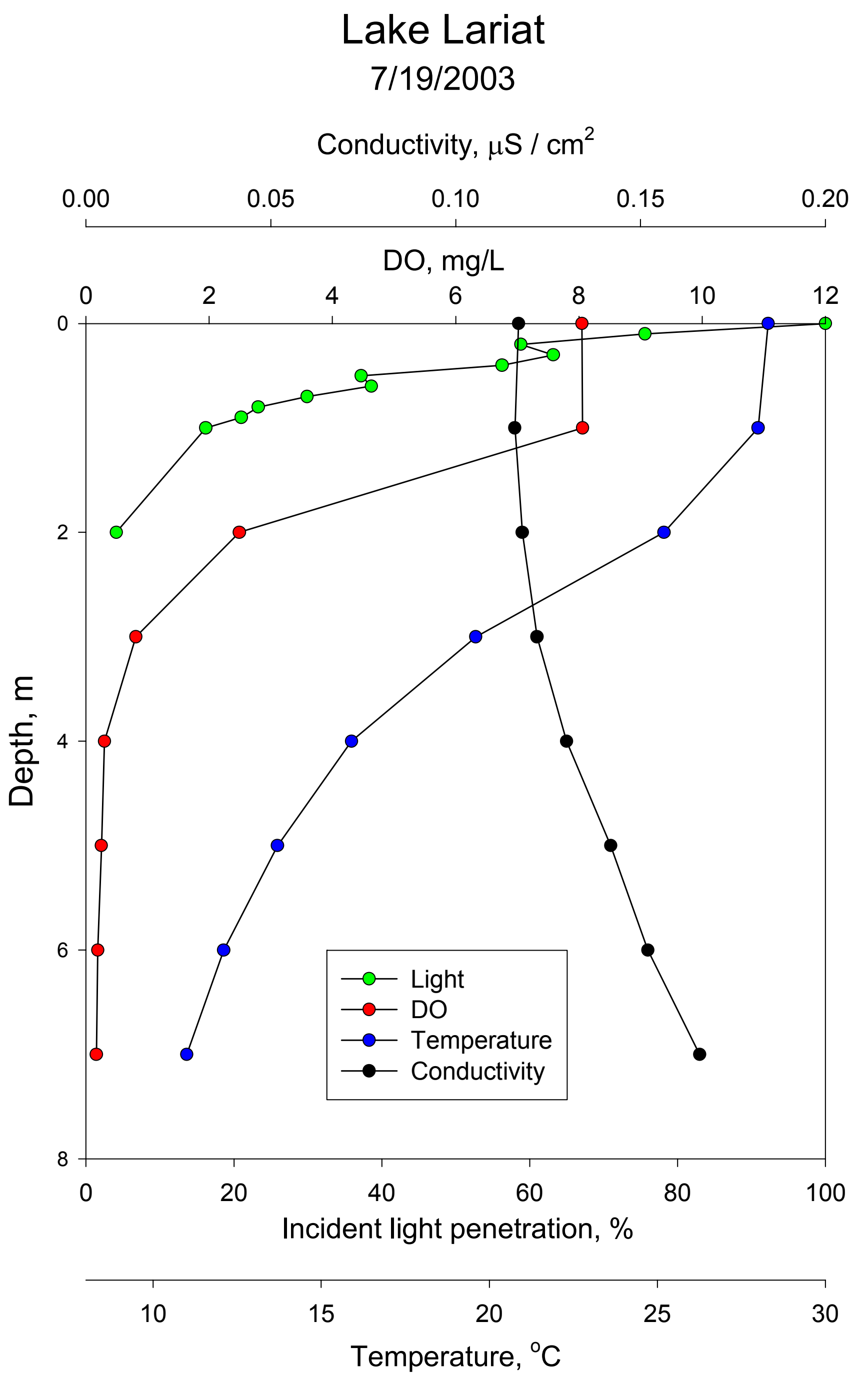


Figure 9A. Depth profile of Lake Lariat, Calvert County, 7/29/2003. Lake Lariat is a first order impoundment of a small coastal stream in a heavily suburban watershed. It is turbid, eutrophic and shallow. However it is strongly stratified and has anoxic bottom water in the summer.

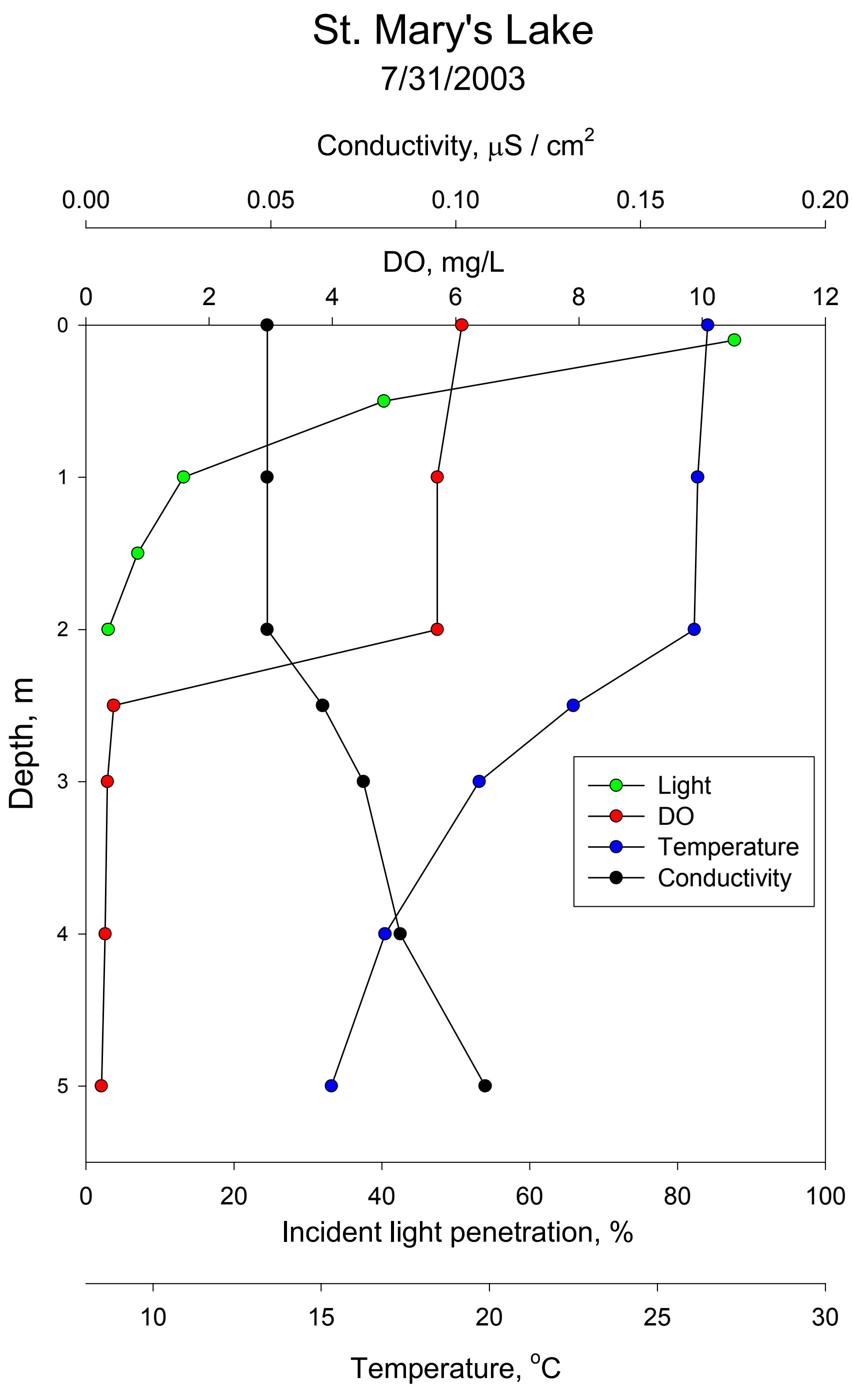


Figure 9B. Depth profile of St. Mary's Lake, St. Mary's County, 7/31/2003. The lake is a first order impoundment of a small river in a protected watershed. It is turbid, highly colored with DOC and has intermittently low pH. It is shallow, but strongly stratified with anoxic bottom water in summer.

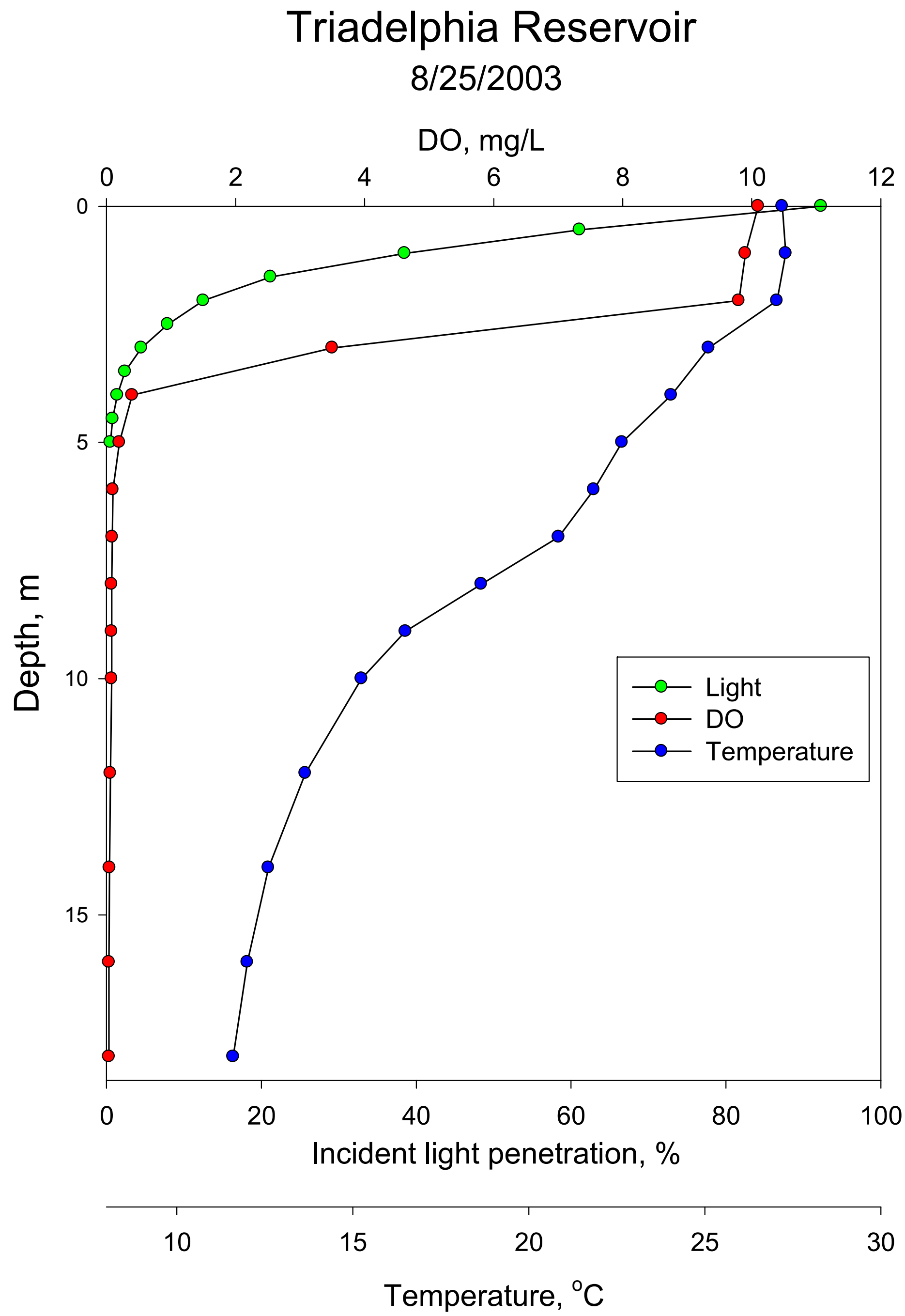


Figure 9C. Depth profile of Triadelphia Reservoir, 8/25/2003. Triadelphia is a large water supply impoundment of the Patuxent River. It is relatively deep reservoir, with a large mixed used upstream watershed. The area around the reservoir is protected forest buffer. On the sampling date, the water was stratified with a sharp oxycline but diffuse thermocline, and anoxic bottom waters.

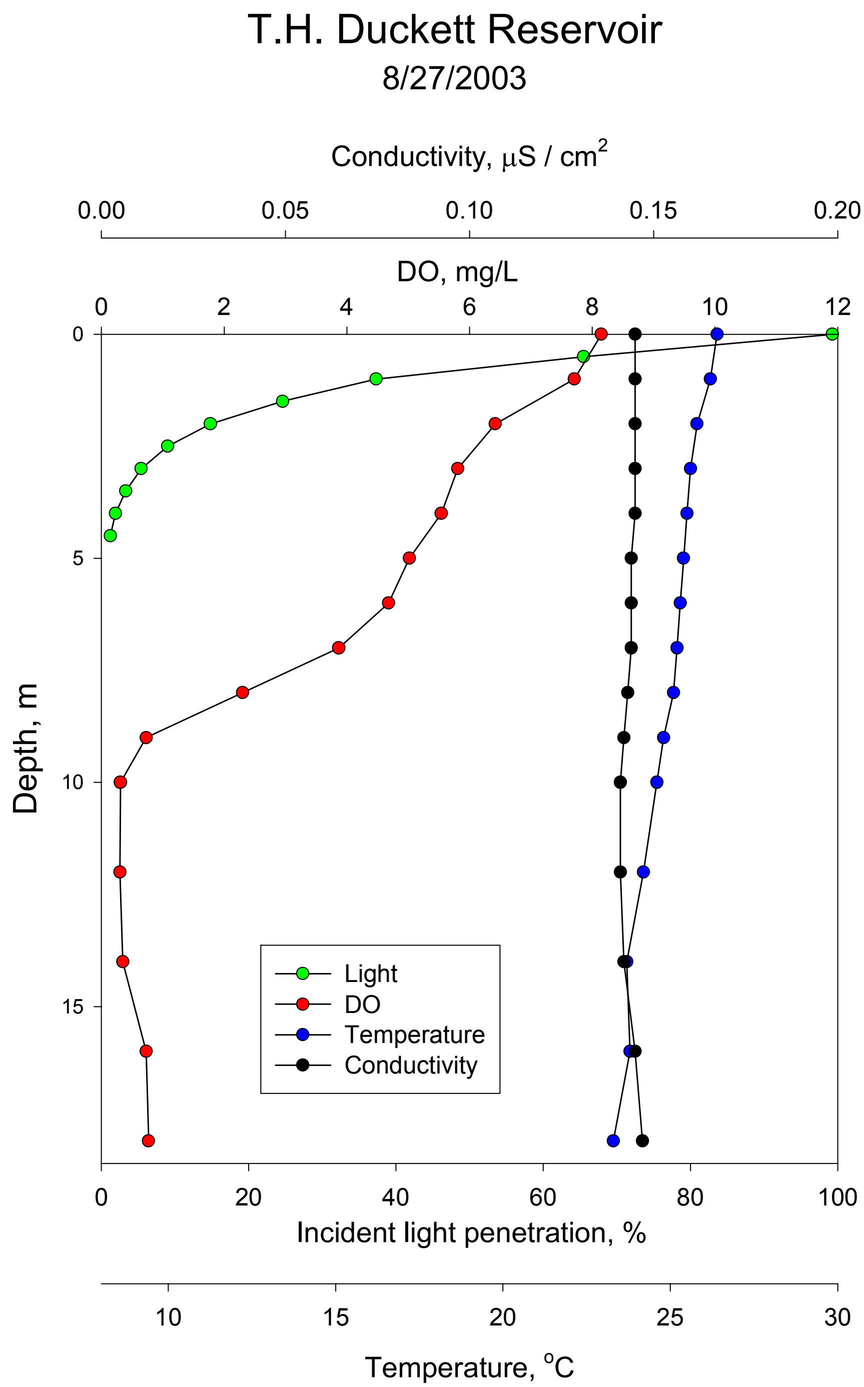


Figure 9D. Depth profile of T.H. Duckett Reservoir, 8/27/2003. Duckett (Rocky Gorge) is also a large water supply impoundment of the Patuxent River, below Triadelphia Reservoir. It is relatively deep with a large mixed-use upstream watershed, although the area around the reservoir is protected forest buffer. The bottom waters are anoxic, but there is little or no temperature or conductivity stratification in this high-flow system.

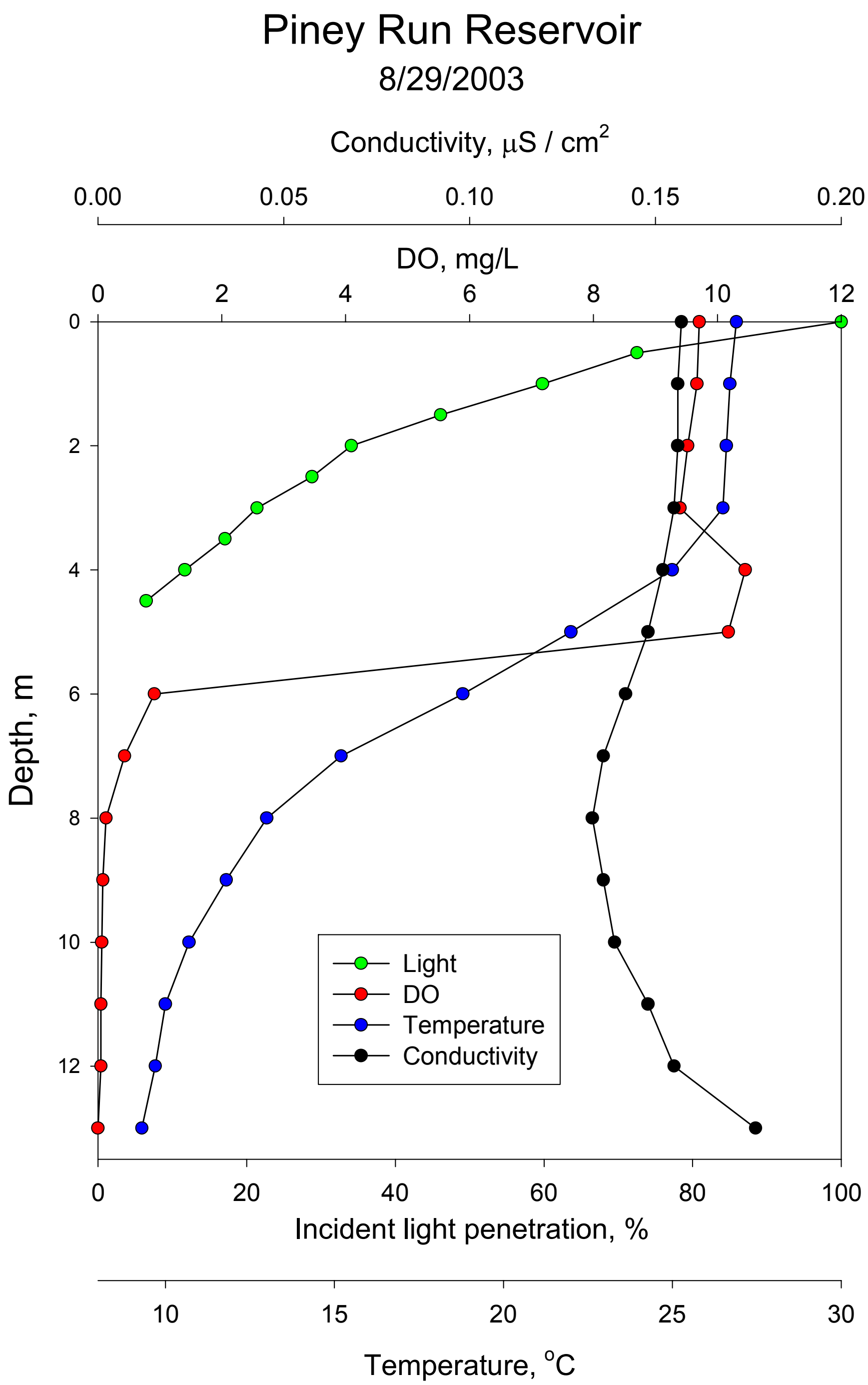


Figure 9E. Depth profile of Piney Run Reservoir, 8/29/2003. Piney Run is in the Patapsco watershed. It is one the clearer reservoirs examined, relatively deep, and strongly stratified with anoxic bottom waters.

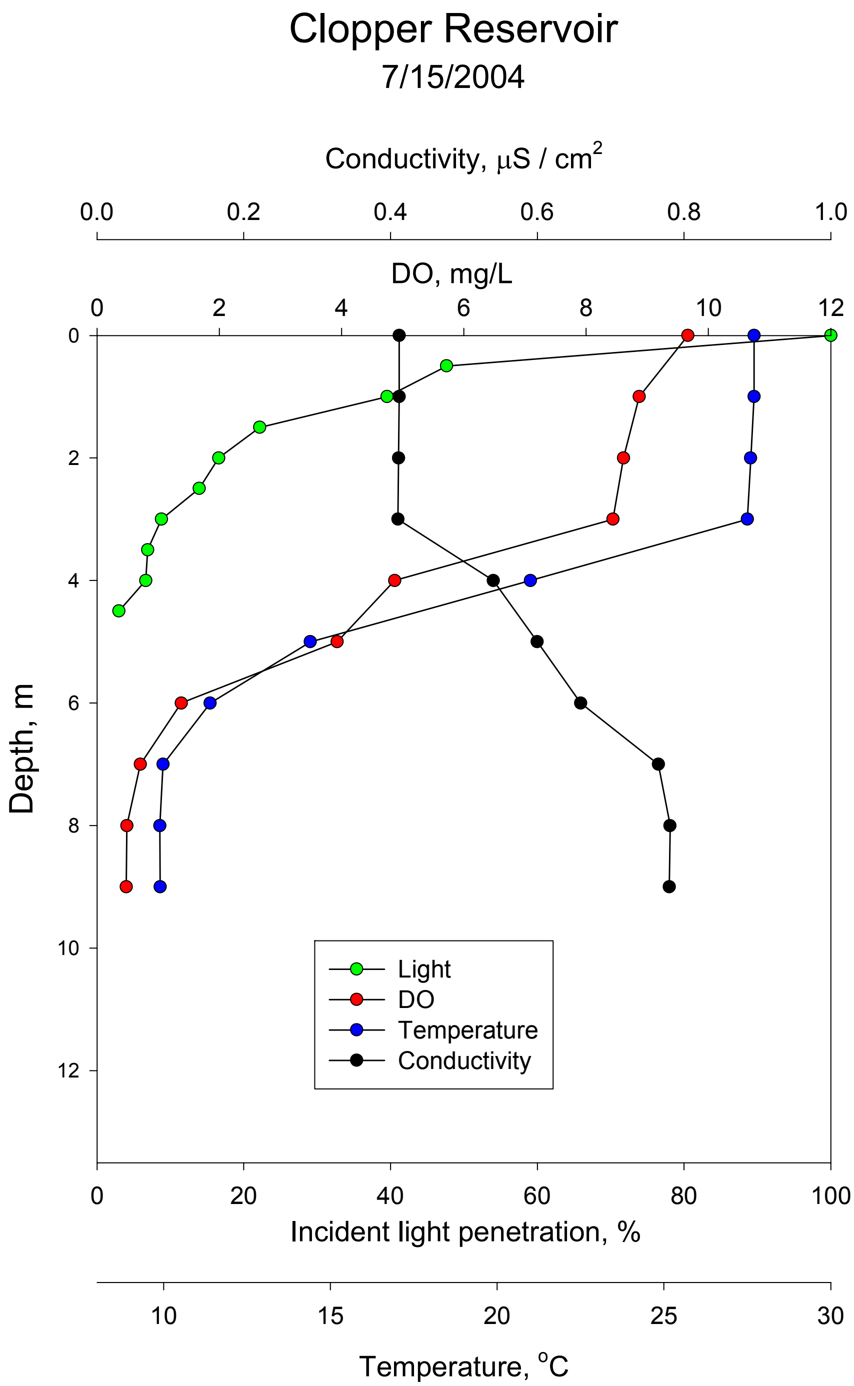


Figure 9F. Depth profile of Clopper Reservoir, 7/15/2004. This suburban reservoir in Montgomery County is eutrophic, highly stratified with anoxic bottom waters.

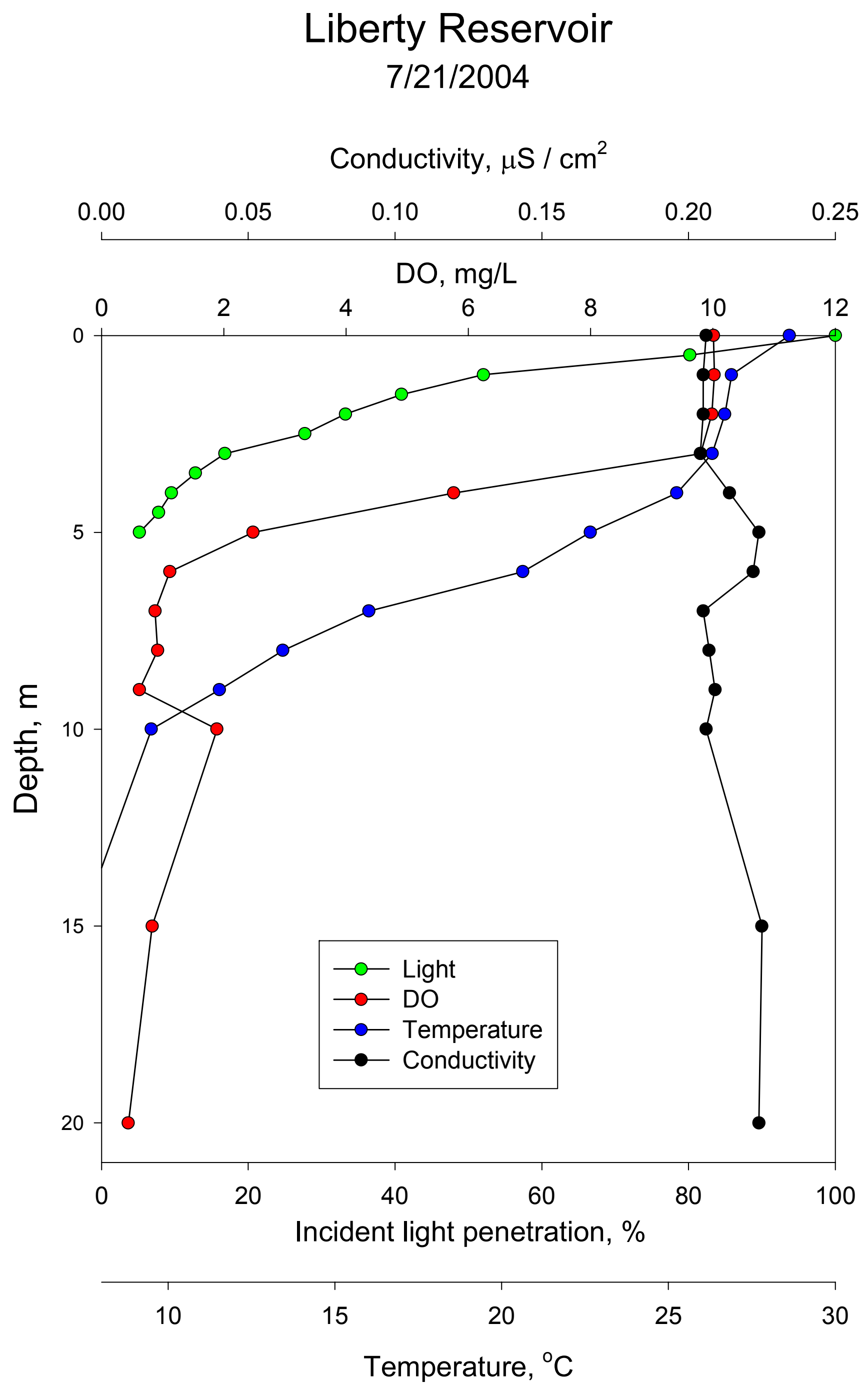


Figure 9G. Depth profile of Liberty Reservoir, 7/21/2004. This large reservoir west of Baltimore is relatively deep, and highly stratified with anoxic bottom waters. Based on light penetration data on this date, it is one of the clearer reservoirs sampled.

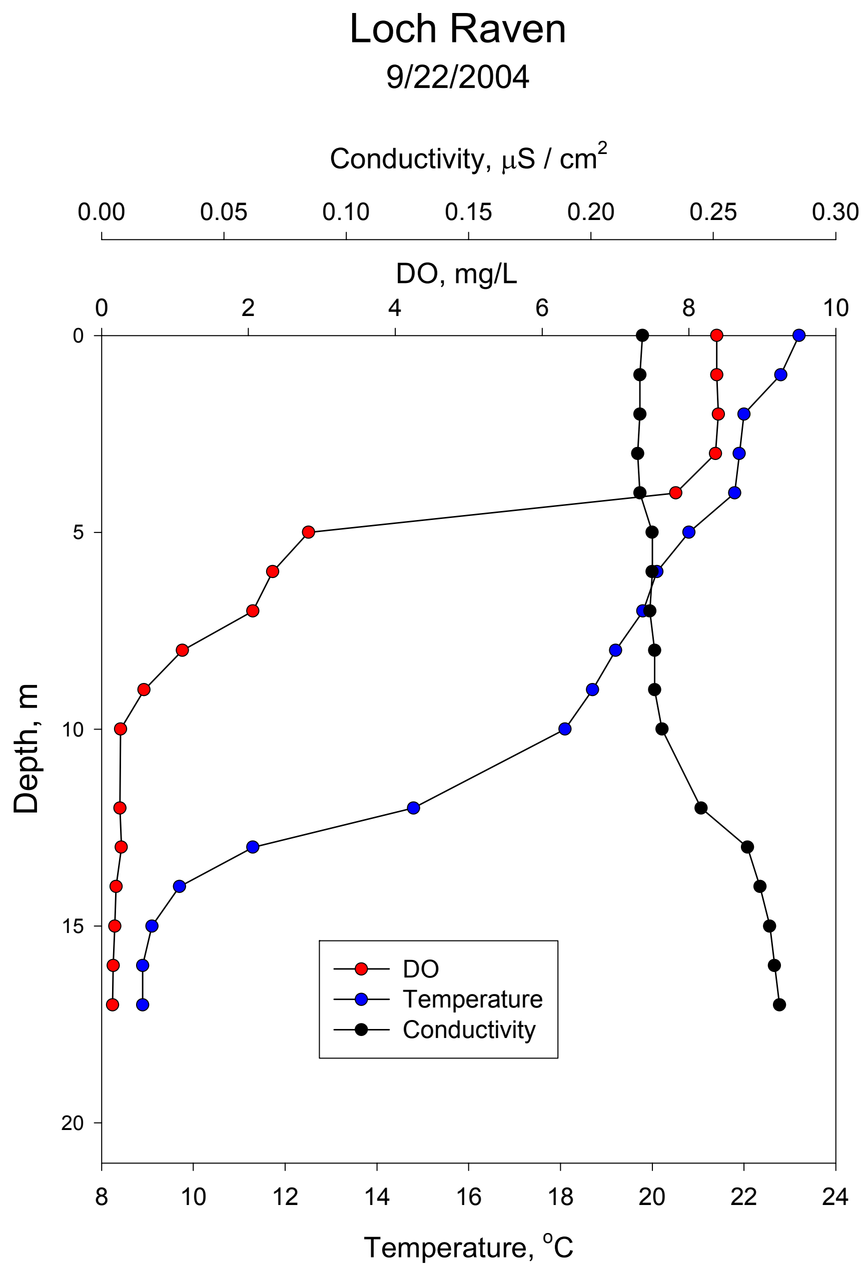


Figure 9H. Depth profile of Loch Raven Reservoir, 9/22/2004. Loch Raven is just north of Baltimore on the Patapsco. The reservoir is strongly stratified with anoxic bottom waters, although on this date, the difference in temperature and oxygen profiles suggests it is near fall overturn.

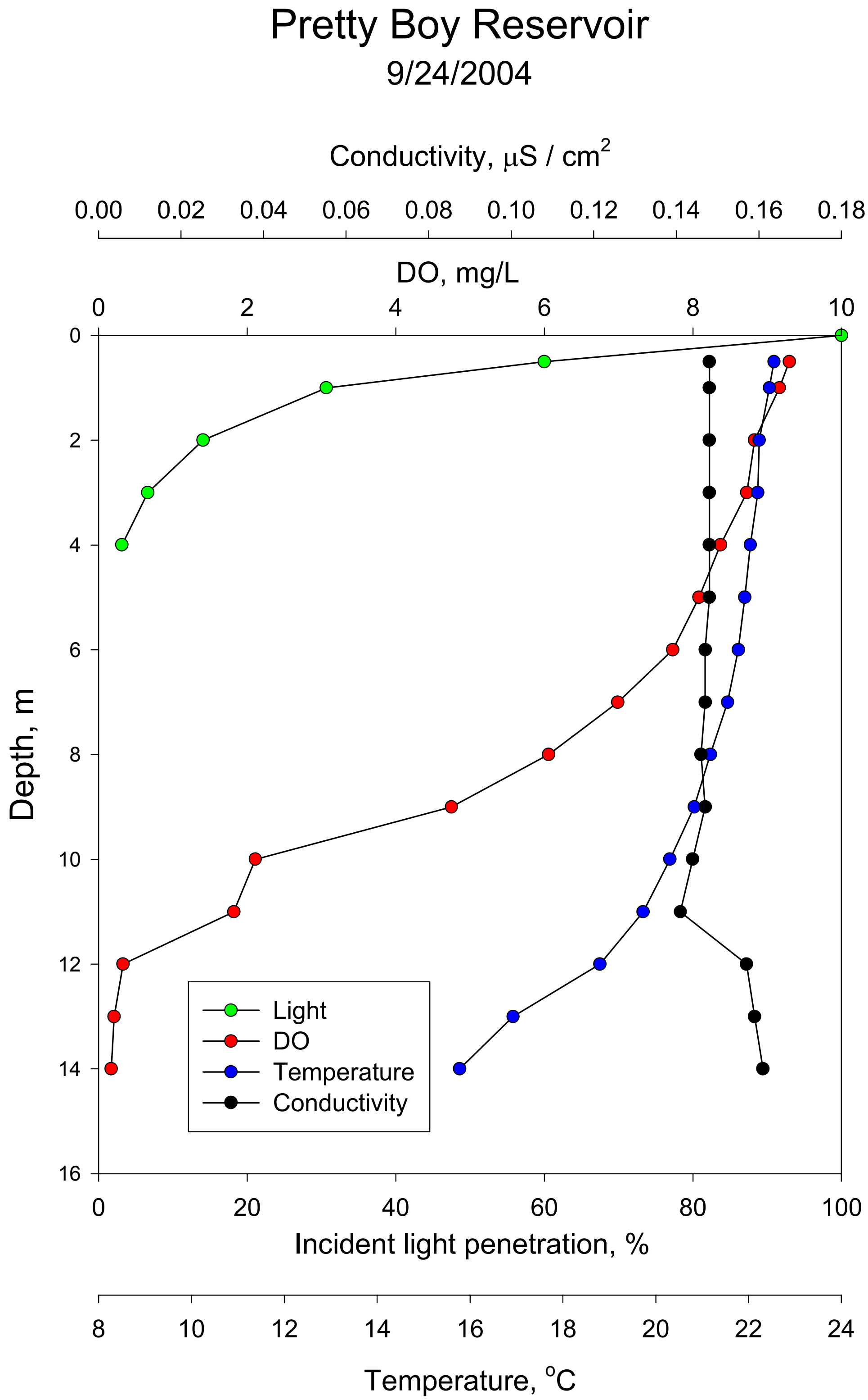


Figure 9I. Depth profile of Prettyboy Reservoir, 9/24/2004. Prettyboy Reservoir is an impoundment in the Gunpowder River Watershed in northwestern Baltimore County, Maryland.

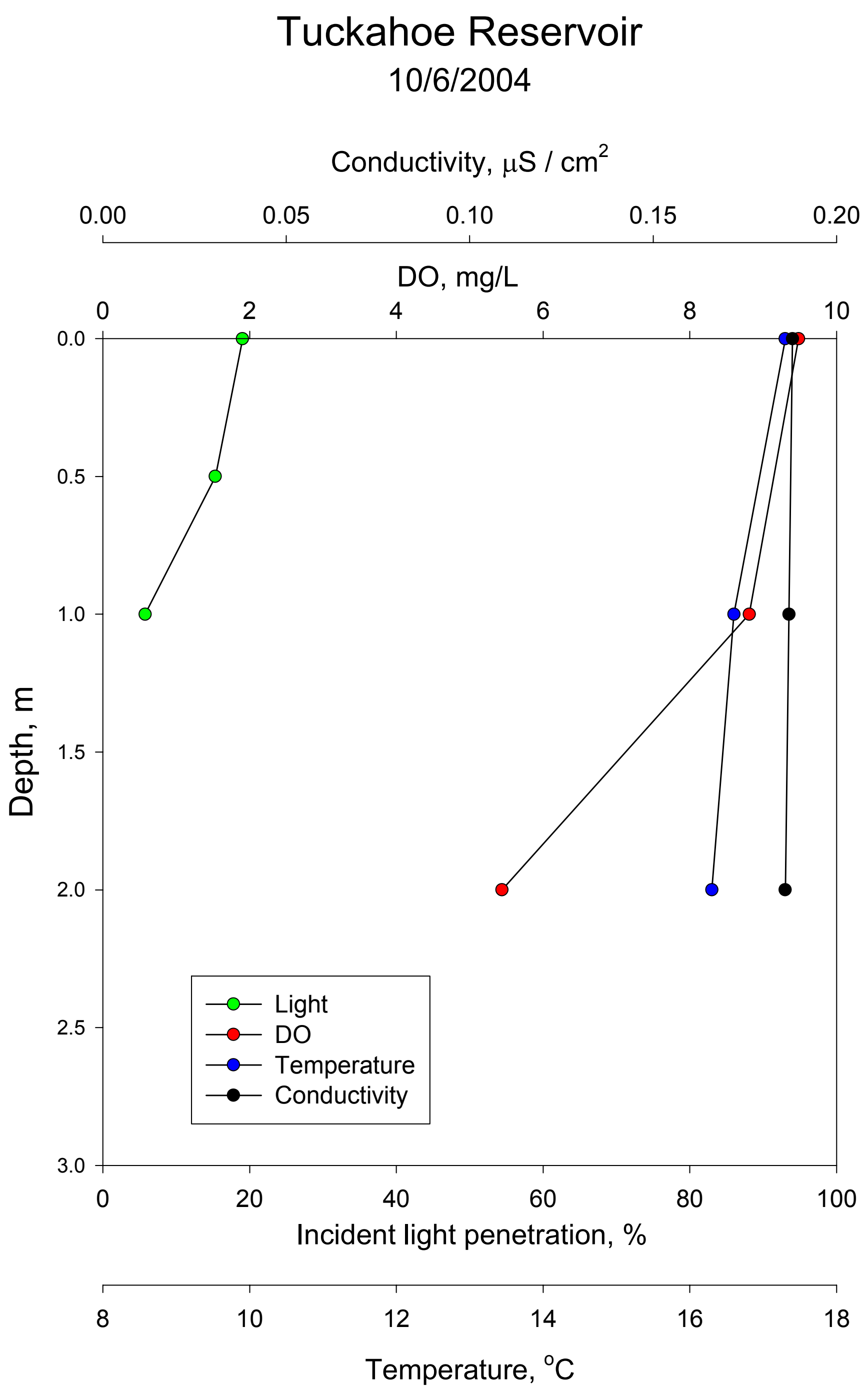


Figure 9J. Depth profile of Tuckahoe Reservoir, 10/6/2004. Tuckahoe is a small, very shallow impoundment on the Eastern Shore.

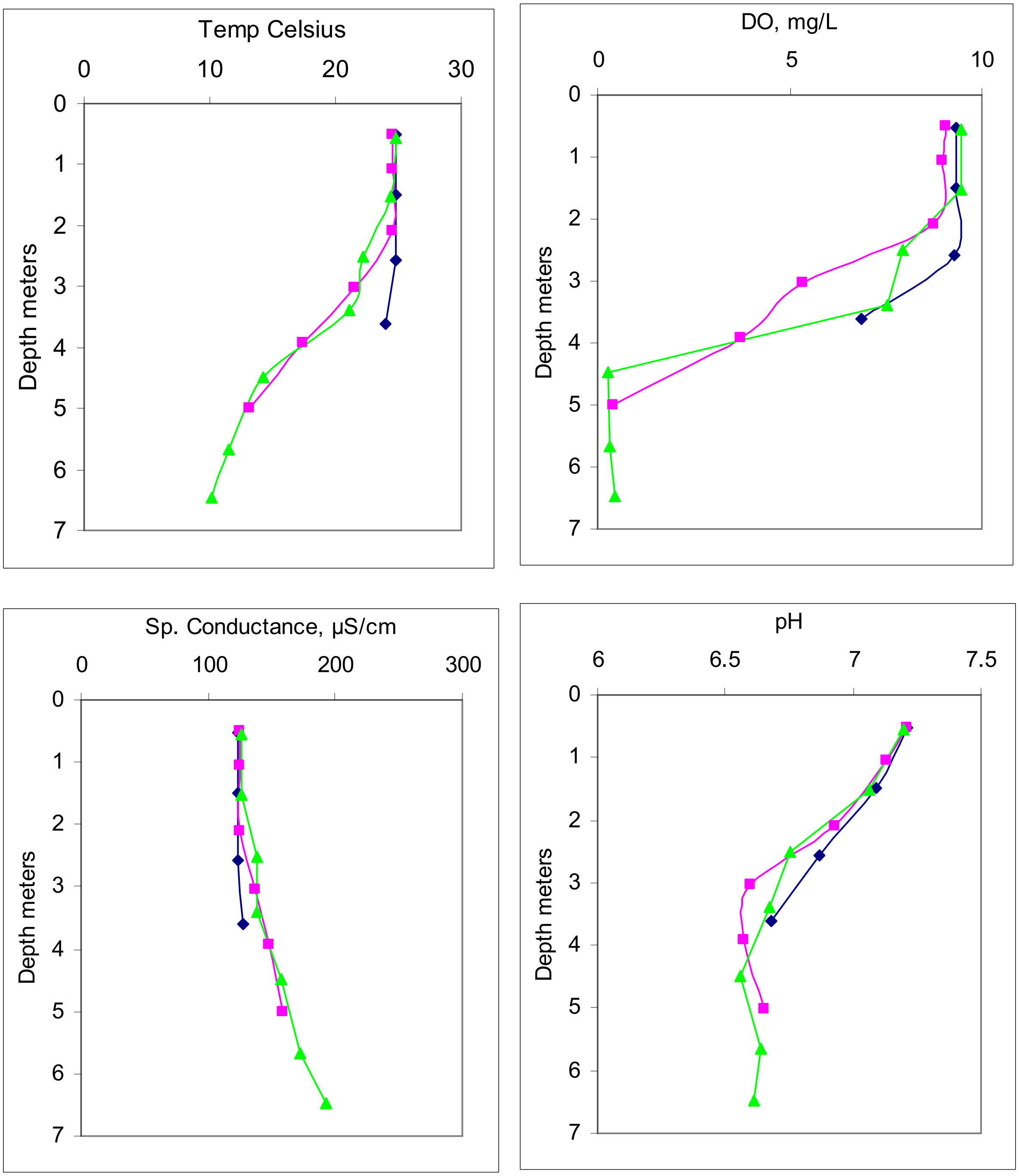


Figure 10A. Water column depth profiles at three sites in Piney Reservoir, 7/14/2004.

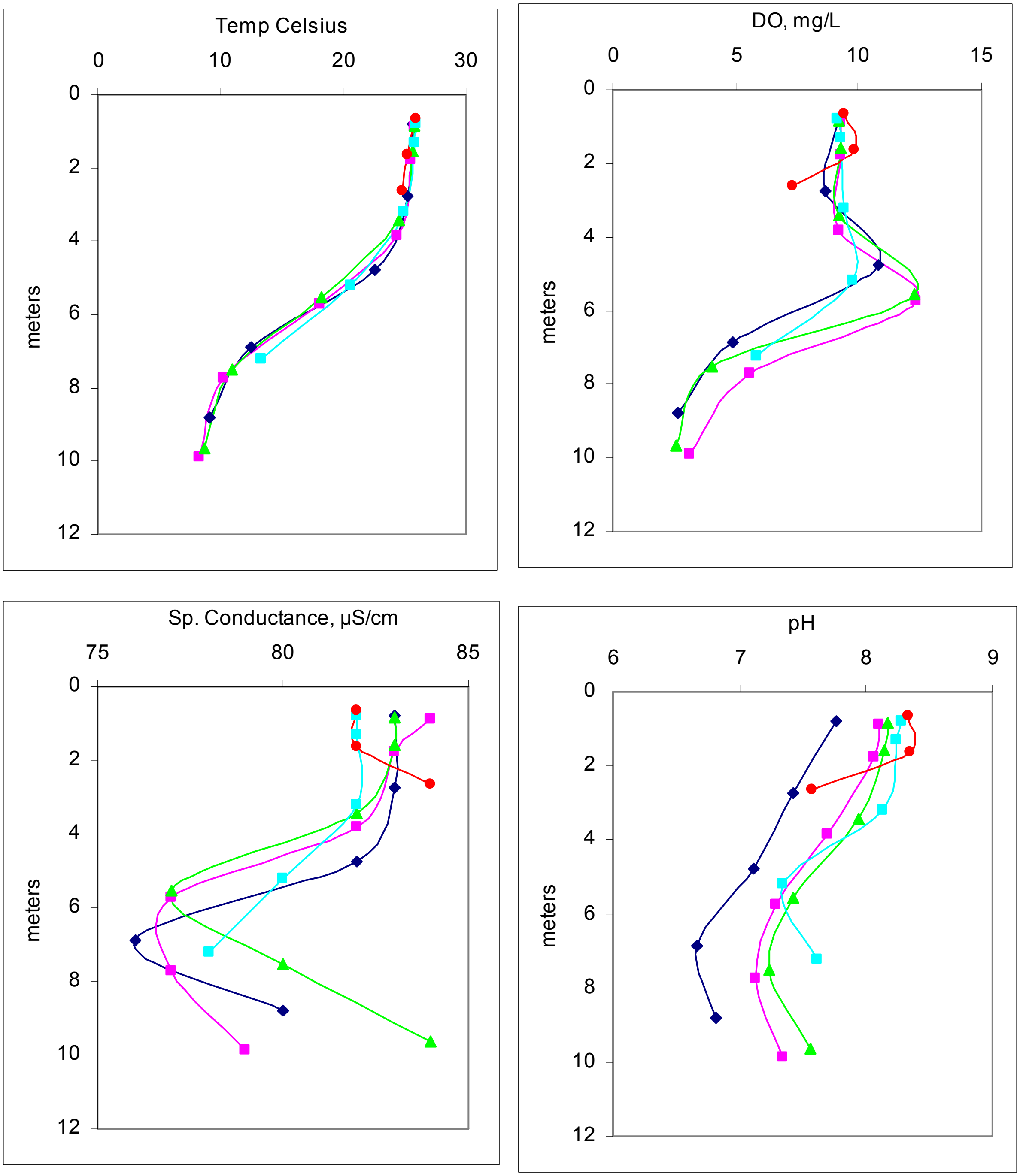


Figure 10B. Water column depth profiles at five sites in Rocky Gap Reservoir, 7/22/2004.

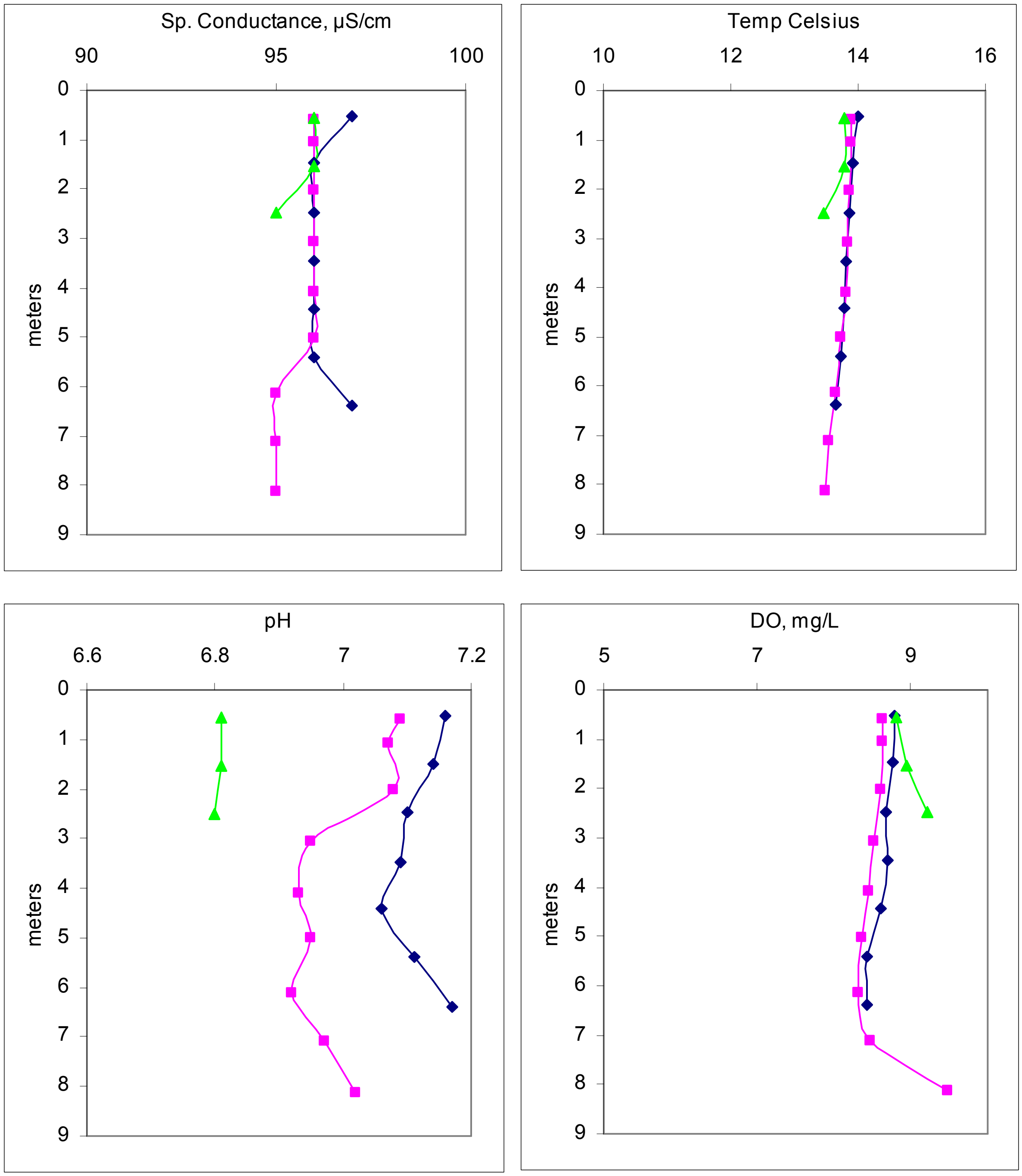


Figure 10C. Water column depth profiles at three sites in Savage Reservoir, 11/3/2004.

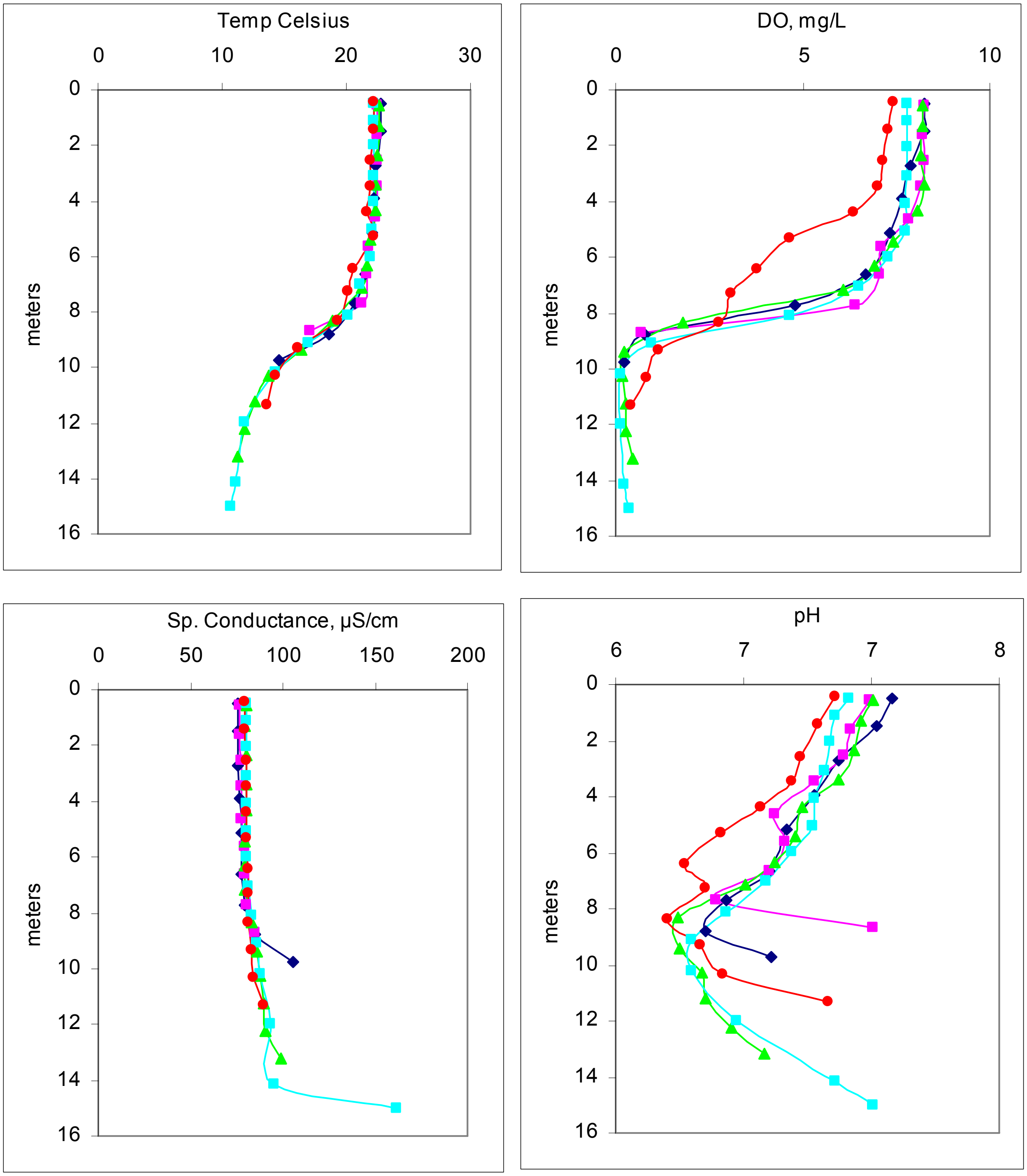


Figure 10D. Water column depth profiles at five sites in Deep Creek Lake, 8/11/2004.

***Water chemistry*.** Water chemistry for the 10 eastern reservoirs in 2003-2004, including Hg and MeHg concentrations is given in Tables 9 and 11. Data for the 4 western reservoirs are in Tables 10 and 12.

Filtered and unfiltered mercury concentrations in surface waters varied substantially across the reservoirs. Mercury concentrations tended to be highest in Coastal Plain and Piedmont reservoirs and lowest in the western reservoirs (Figure 11). Many reservoirs contained quite low total Hg concentrations - below 1 ng/L in some reservoirs, while a few contained 2-6 ng total Hg/L. We found very high total Hg levels in T. Howard Duckett reservoir. Concentrations in the 15 and 45 ng/L range (filtered and unfiltered) were observed during this study. Svensdottir et al. also measured high concentrations, between 15 and 20 ng/L, in 2000-2001. A further examination of Hg in Duckett is probably warranted. Filterable Hg concentrations were generally, but not always, higher in bottom waters than in surface waters.

Among MD reservoirs, water chemistry parameters significantly related to dissolved Hg included chloride, dissolved organic carbon and sulfate (Figure 12). These relationships were examined using the average concentration of the variables for each reservoir, from 2000-2005. The data set used for this graphic can be found in Tables 18 and 19. Dissolved Hg concentrations were positively related to DOC and chloride, and negatively related to sulfate. Both chloride and DOC can be strong ligands for Hg and may help to hold it in solution. However, Cl concentrations were also strongly correlated with land use. Reservoirs with a higher fraction of developed land had both higher Hg and Cl concentrations.

Surface water total MeHg concentrations ranged from near our detection limit of ~0.05 ng/L up to more than 2 ng/L (Figure 13), which is considered a substantial concentration for a natural water not contaminated by a point source of Hg. Methylmercury levels were highest by a substantial margin in two of the Coastal Plain reservoirs, St. Mary's Lake and Lake Lariat.

One way to examine the conversion of inorganic Hg to MeHg across the reservoirs is to normalize MeHg to the Hg concentration. Figure 14 shows the percent MeHg for each lake, where:

$$\% \text{ MeHg} = (\text{MeHg}/\text{Hg}) \times 100$$

A substantial percent of the surface water Hg could be found as MeHg in the four eastern reservoirs, particularly those with anoxic bottom waters, and the two small, eutrophic Coastal Plain reservoirs (St. Mary's Lake and Lake Lariat). The percent of dissolved Hg as MeHg (%MeHg) was generally lower in the four western reservoirs, except in the anoxic bottom waters of Deep Creek Lake. In general, the %MeHg in eastern Maryland reservoirs was quite high.

Table 9. Water column Hg and MeHg concentration data for eastern reservoirs from CBL/SERC 2003-2004. The standard deviations presented are for analytical duplicates.

Reservoir	Date	Depth (m)	HgT (ng/L) Unfiltered	std	HgT (ng/L) Filtered	std	MeHg (pg/L) Unfiltered	std	MeHg (pg/L) Filtered	std	%MeHg unfiltered	%MeHg filtered
Clopper	7/15/2004	0.5	2.60		0.66		0.22		0.12		8.6	18.4
Clopper	7/15/2004	6	1.46		0.51		0.20		0.14		13.7	26.4
Duckett Res.	8/27/2003	1	18.04	0.43	35.02	1.66	0.18	0.01	0.13	0.03	1.0	0.4
Duckett Res.	8/27/2003	18	46.31	0.04	35.73	0.08	0.07	0.04	0.04	0.02	0.1	0.1
Lake Lariat	7/29/2003	1	2.74		1.89		1.59	0.05	1.10	0.02	58.0	58.1
Lake Lariat	7/29/2003	7	1.97		1.58		0.35	0.08	0.25	0.01	18.0	16.1
Liberty	7/21/2004	0.5	0.51	0.03	0.55		0.14		0.16		28.1	29.5
Liberty	7/21/2004	20	1.35	0.07	0.48		0.18	0.08	0.10		13.1	21.8
Loch Raven	9/22/2004	0.5	0.60		0.45		0.37		0.15		61.6	32.6
Loch Raven	9/22/2004	16	0.81		0.87		0.40		0.09	0.01	49.2	10.7
Piney Run Lake	8/29/2003	2	0.70	0.04	0.43	0.00	0.00		0.04		0.0	8.7
Piney Run Lake	8/29/2003	13	1.84	0.15	0.78	0.24	0.23	0.23	0.20	0.01	12.5	25.4
Pretty Boy	9/24/2004	0.5	1.15		0.47		0.21	0.00	0.09		18.4	20.1
Pretty Boy	9/24/2004	13	2.08	0.04	0.47		0.12		0.07		5.9	15.7
St Mary's Lake	7/31/2003	1	6.17		0.84	0.15	0.34		0.21	0.01	5.5	25.1
St Mary's Lake	7/31/2003	5	3.26		1.85		2.79	0.17	1.92	0.14	85.5	103.8
Triadelphia Res.	8/25/2003	0.5	6.71	0.86	0.38	0.07	0.04	0.00	0.09	0.01	0.6	24.7
Triadelphia Res.	8/25/2003	12	1.52	0.02	0.61	0.21	0.20	0.00	0.05	0.03	12.9	8.8
Tuckahoe	10/6/2004	flooded	0.76		0.57	0.01	0.13		0.10		17.8	16.6
Tuckahoe	10/6/2004	open water	1.15		0.56	0.01	0.10		0.04		8.6	7.5

Table 10. Water column Hg and MeHg concentration data for western reservoirs 2004-2005 from AL. Detailed information can be found in Castro 2006.

Reservoir	Date	Depth (m)	HgT (ng/L) Unfiltered	HgT (ng/L) Filtered	MeHg (pg/L) Unfiltered	%MeHg unfiltered
Piney Res	7/14/04, 7/20/05	surf	0.84	0.54	0.038	4.5
Piney Res	7/14/04, 7/20/05	bottom	1.19	0.71	0.107	9.0
Rocky Gap	10/22/03, 7/23/04	surf	0.63	0.45	0.019	3.0
Rocky Gap	10/22/03, 7/23/04	bottom	0.80	0.42	0.022	2.7
Savage	8/4/04, 8/17/05	surf	0.55	0.34	0.069	12.6
Savage	8/4/04, 8/17/05	bottom	0.67	0.38	0.058	8.7
Deep Creek	8/11/04, 9/14/05	surf	0.33	0.21	0.034	10.3
Deep Creek	8/11/04, 9/14/05	bottom	0.75	0.39	0.360	48.3

Table 11 . Water chemistry data for eastern reservoirs for 2003-2004 from CBL/SERC. Standard errors represent analytical duplicates. Missing values were not determined.

Reservoir	Date	Depth (m)	pH	TSS (mg/L)	DOC	PC (mg/L)	PN (mg/L)	Chloride (mg/l)	std	Nitrite (mg/L)	std	Nitrate (mg/l)	std
Clopper	7/15/2004	0.5	7.95	5.058	10.35	0.464	0.076	88.9	0.4			0.45	0.04
Clopper	7/15/2004	6	7.43	1.404	9.51	1.350	0.175	160.3	11.1			0.30	1.18
Duckett Res.	8/27/2003	1	7.10	9.760	3.57								
Duckett Res.	8/27/2003	18	6.98	8.600	1.47								
Lake Lariat	7/29/2003	1	6.45	4.900	5.33								
Lake Lariat	7/29/2003	7	6.52	6.900	3.00								
Liberty	7/21/2004	0.5		1.94	16.12	0.612	0.066	26.1	0.0	0.06	0.01	2.05	0.71
Liberty	7/21/2004	20		2.54	21.17	0.507	0.054	30.1	0.2	0.07	0.00	2.37	0.31
Loch Raven	9/22/2004	0.5	8.05	0.00	9.20	0.539	0.076	26.2		0.06		1.23	
Loch Raven	9/22/2004	16	7.45	0.00	13.66	0.387	0.058	30.4				1.18	
Piney Run Lake	8/29/2003	2	6.60	6.80	4.00								
Piney Run Lake	8/29/2003	13	7.09	11.00	1.30								
Pretty Boy	9/24/2004	0.5	7.65	0.00	14.85	1.160	0.189	17.3		0.05		1.51	
Pretty Boy	9/24/2004	13	7.61	0.03	23.02	1.560	0.207	16.8				1.54	
St Mary's Lake	7/31/2003	1	4.18	18.04	9.56								
St Mary's Lake	7/31/2003	5	5.50	16.40	13.64								
Triadelphia Res.	8/25/2003	0.5	6.61	30.84	2.98								
Triadelphia Res.	8/25/2003	12	6.03	12.64	3.77								
Tuckahoe	10/6/2004	flooded	7.35	0.01	11.23	0.684	0.095	15.1		0.05		2.17	
Tuckahoe	10/6/2004	open water	7.64	0.00	12.34	0.755	0.110	15.0	0.1	0.05	0.00	3.68	0.00

Table 11. continued. Water chemistry data for eastern reservoirs, 2003-2004 from CBL/SERC. Standard errors represent analytical duplicates. Missing values were not determined.

Reservoir	Date	Sulfate (mg/l) avg	Sulfate (mg/l) std	Sulfide (mg/L)	10% Incident light depth (m)	Chla, total (ug/L)	Phaeo (ug/L)	Active Chl (ug/L)	Bottom DO (mg/L)	DO (mg/L)	Temp C	Conductivity (uS/cm)
Clopper	7/15/2004	3.00	0.36	BDL	2.8	2.83	1.63	2.02		8.87	27.7	0.412
Clopper	7/15/2004	2.65	1.63	BDL		5.77	5.67	2.94	0.48	1.38	11.4	0.659
Duckett Res.	8/27/2003	6.61		0.017	2.25					7.71	26.2	0.145
Duckett Res.	8/27/2003	6.35		0.019					0.77	0.77	23.3	0.147
Lake Lariat	7/29/2003	6.78		BDL	1.5					8.06	28.0	0.116
Lake Lariat	7/29/2003	6.04		0.079					0.17	0.17	11.0	0.166
Liberty	7/21/2004	2.68	0.60		3.9	6.57	2.03	5.55		10.02	26.9	0.205
Liberty	7/21/2004	2.64	0.16			2.38	2.68	1.04	0.48	0.44	7.4	0.224
Loch Raven	9/22/2004	2.73		BDL		5.56	0.68	5.22		8.38	22.8	0.220
Loch Raven	9/22/2004	2.70		BDL		1.82	1.05	1.30	0.15	0.16	8.9	0.275
Piney Run Lake	8/29/2003	5.06		BDL	4.25					9.52	26.6	0.156
Piney Run Lake	8/29/2003	UND		0.127					0	0.00	9.3	0.177
Pretty Boy	9/24/2004	1.82		BDL	2.5	18.44	0.53	18.17		9.17	22.5	0.148
Pretty Boy	9/24/2004	1.78		BDL		6.04	6.87	2.61	0.17	0.21	16.9	0.159
St Mary's Lake	7/31/2003	4.82		BDL	1.25					5.70	26.2	0.049
St Mary's Lake	7/31/2003	0.95		0.023					0.25	0.25	15.3	0.108
Triadelphia Res.	8/25/2003	5.29		BDL	2.25					9.90	27.3	NA
Triadelphia Res.	8/25/2003	5.81		BDL					0.04	0.06	13.7	NA
Tuckahoe	10/6/2004	4.13		0.005		5.04	3.84	3.12	5.44	8.81	16.6	0.187
Tuckahoe	10/6/2004	4.41	0.26	0.01	0.75	13.56	4.26	11.43		NA	NA	NA

Table 12. Water chemistry data for western reservoirs from AL, 2004-2005. Data are averages for dates listed.

Reservoir	Date	Depth (m)	pH	TSS (mg/L)	DOC	Total PC (mg/L)	Total PN (mg/L)	Chloride (mg/l)	Nitrate (mg/l)	Sulfate (mg/l)	Sulfide (mg/L)	DO (mg/L)	Temp C	Conduc-tivity (uS/cm)
Piney Frostburg	7/14/04, 7/20/05	surf	7.13	1.60	3.58	0.66	0.14	21.1	0.91	7.74	0.0043	8.90	25.31	0.133
Piney Frostburg	7/14/04, 7/20/05	bottom	6.83	3.58	3.67	0.86	0.17	22.2	0.87	7.78	0.0053	2.25	15.80	0.166
Rocky Gap	10/22/03, 7/23/04	surf	7.67	1.56	3.77	0.58	0.18	3.2	0.04	9.77	0.0025	7.40	19.89	0.085
Rocky Gap	10/22/03, 7/23/04	bottom	7.34	1.58	3.66	0.57	0.21	3.2	0.06	10.11	0.0054	3.99	11.58	0.087
Savage	8/4/04, 8/17/05	surf	7.74	2.13	1.66	0.21	0.08	1.5	0.55	10.90	0.0026	8.57	23.11	0.101
Savage	8/4/04, 8/17/05	bottom	7.28	1.56	1.72	0.3	0.08	1.7	0.59	10.48	0.0031	3.82	15.07	0.099
Deep Creek	8/11/04, 9/14/05	surf	7.18	1.80	2.44	0.33	0.09	8.6	0.08	12.80	0.0020	7.95	22.65	0.083
Deep Creek	8/11/04, 9/14/05	bottom	6.94	2.67	2.31	0.18	0.08	8.9	0.09	11.49	0.0021	0.44	13.09	0.113

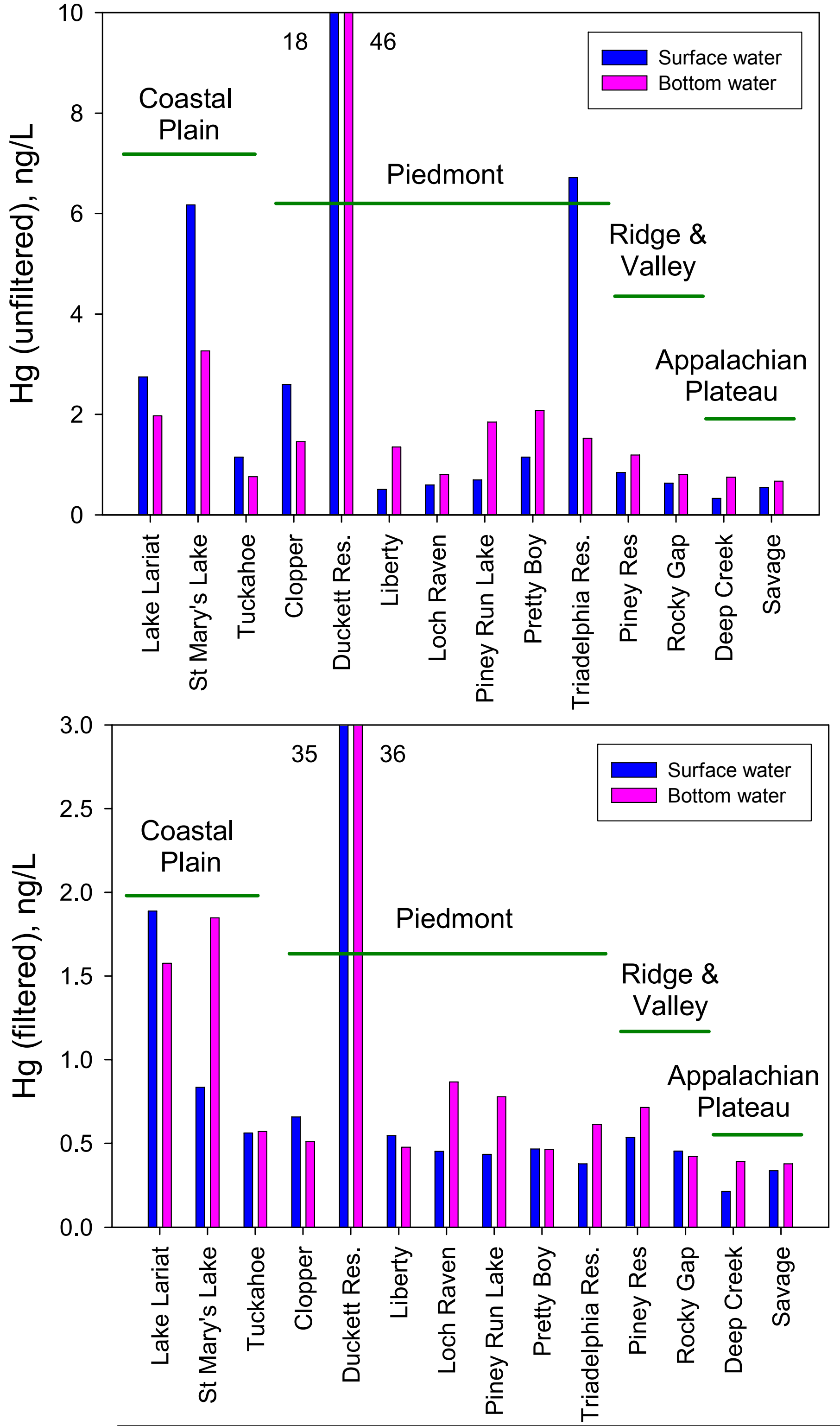
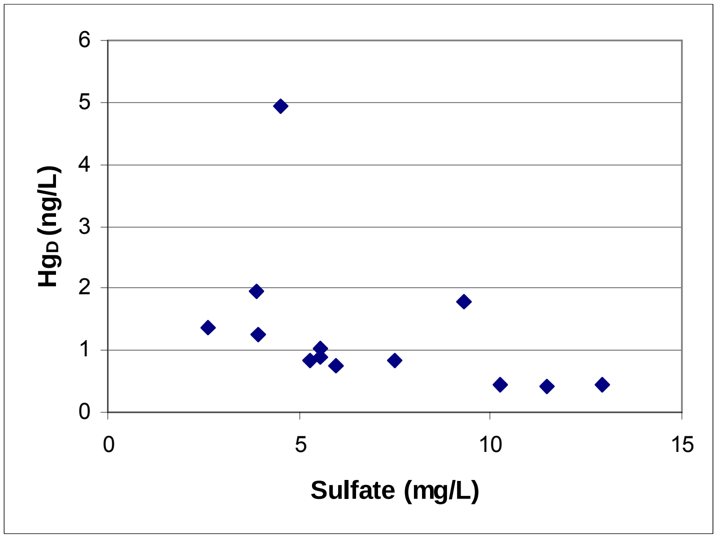
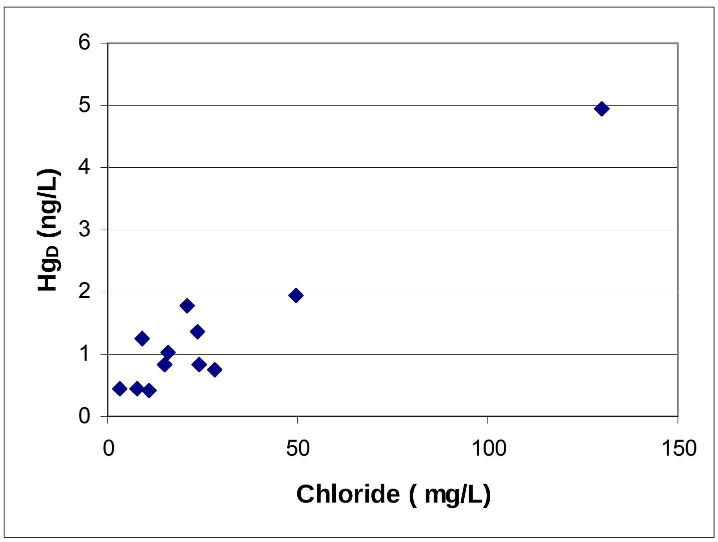


Figure 11. Total (top) and filtered (bottom) Hg concentrations in MD reservoirs, 2003-2005. Blue bars are surface water data, pink bars are bottom waters. Samples were collected at the deepest point in each reservoir. Most bars represent single sample collections.





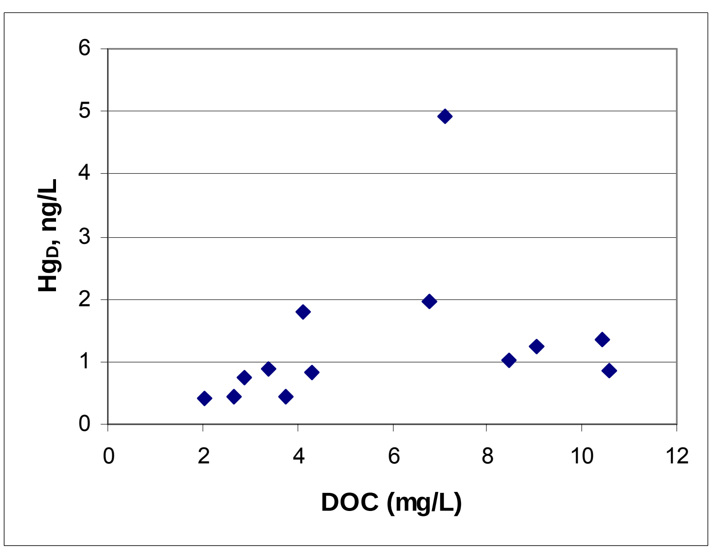


Figure 12. Water chemistry variables significantly related to water column filterable Hg concentrations. Average data 2000-2005 for 13 reservoirs (see Tables 18 and 19); Duckett was excluded because of anomalously high Hg levels. Note that correlations were examined using log transformed variables, in order to normalize data distributions. Statistics for all correlations are in Appendix 1.

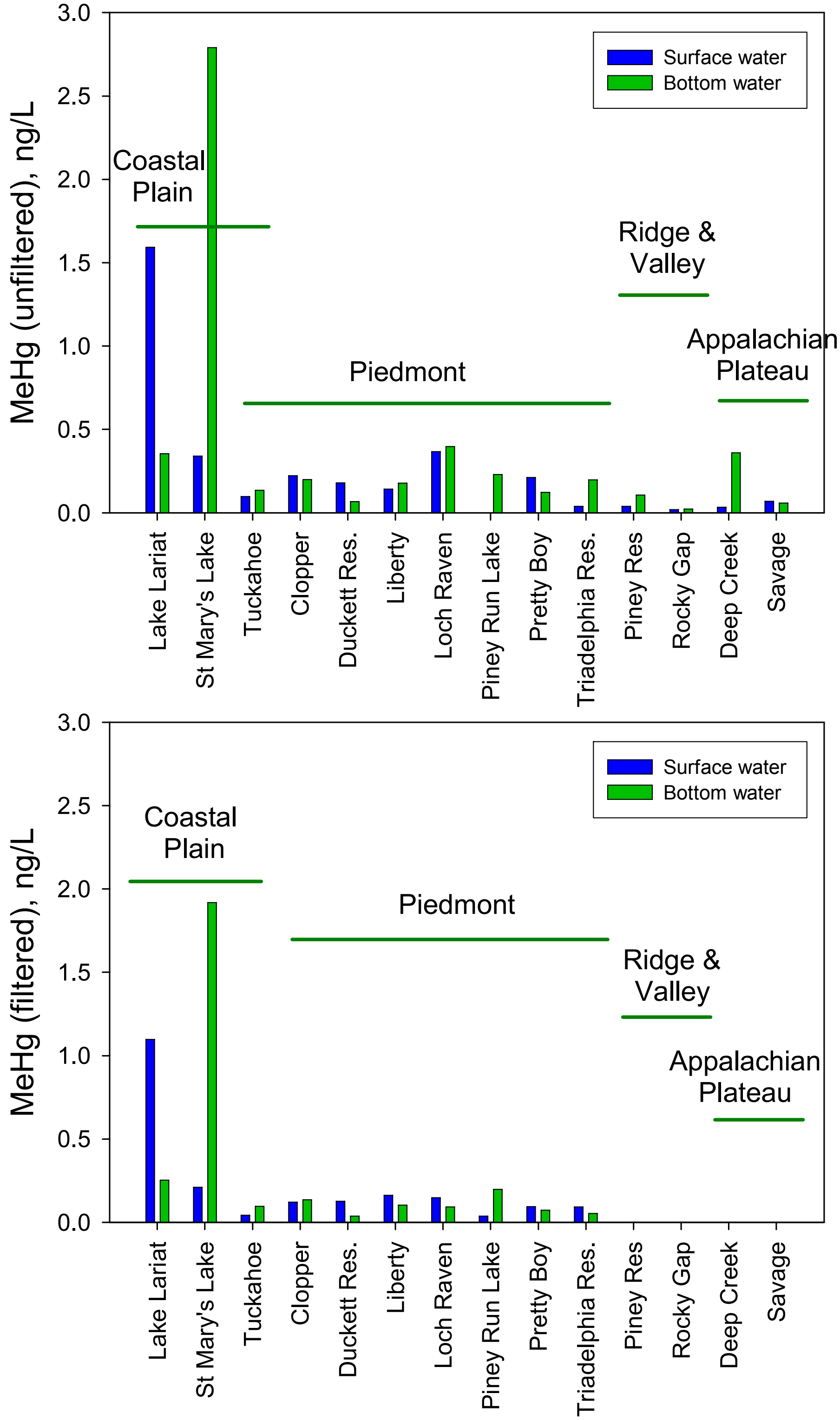


Figure 13. Total (top) and filtered (bottom) MeHg concentrations in MD reservoirs, 2003-2005. Blue bars are surface water data, green bars are bottom waters. Samples were collected at the deepest point in each reservoir. Most bars represent single sample collections. Filtered MeHg data were not collected for the western reservoirs.

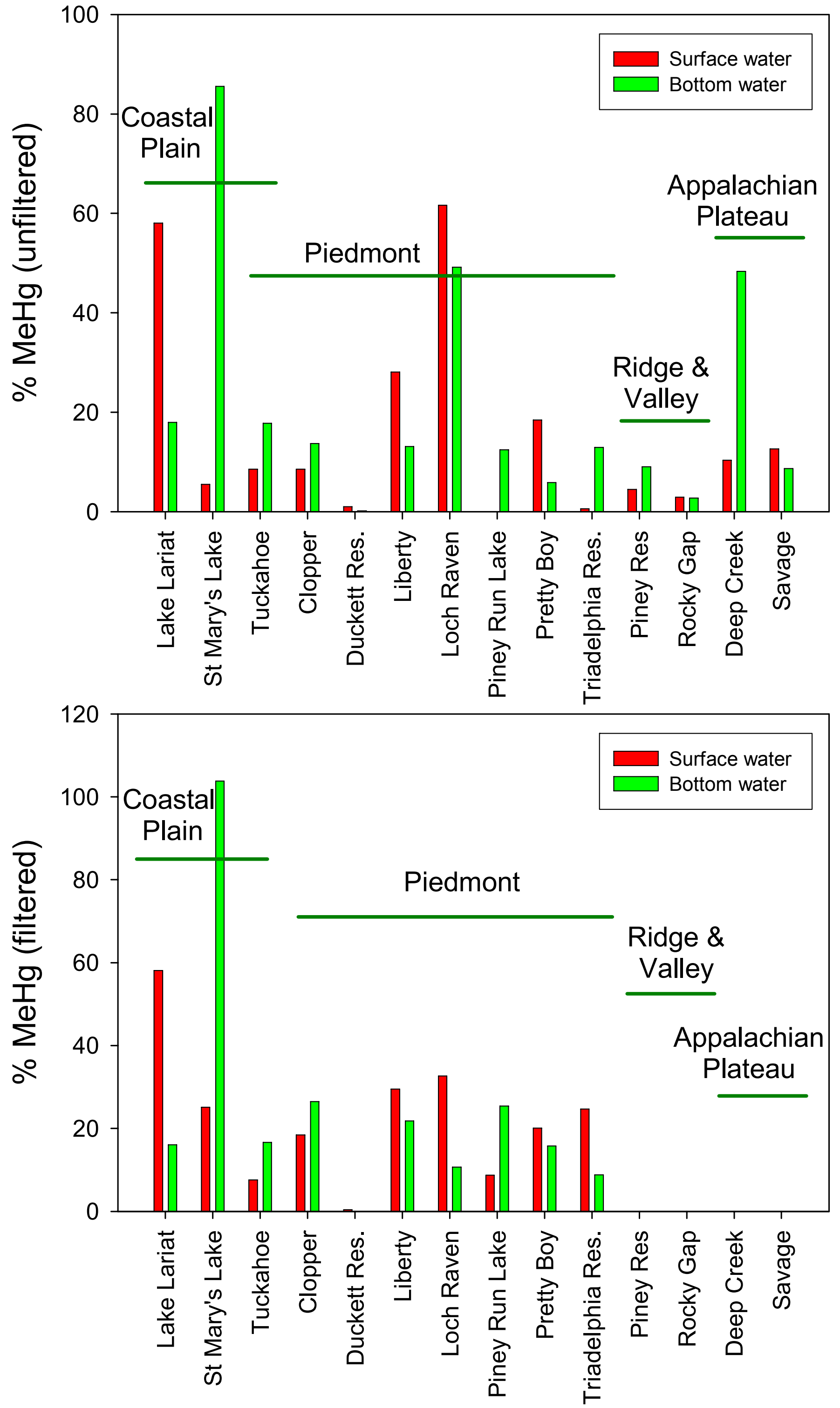


Figure 14. MeHg as a percentage of total Hg, unfiltered (top) and filtered (bottom) for MD reservoirs, 2003-2005. Red bars are surface water, green bars are bottom waters. Filtered MeHg data were not collected for the western reservoirs.

One good predictor of high %MeHg was dissolved oxygen < 1 mg/L (Figure 15). Low DO can enhance MeHg production in sediments and MeHg flux from sediments to overlying waters. MeHg production may also occur in anoxic bottom waters.

Two other variables, pH and sulfate, were significantly correlated with %MeHg (filterable) among the reservoirs (Figure 16). These relationships were examined using the average concentration of the variables for each reservoir from 2000-2005.

Most of the reservoirs examined were circumneutral, although pH was lower in Coastal Plain reservoirs Lake Lariat and St. Mary's Lake (Figure 17).We have observed wide swings in pH (4 to 10) at St. Mary's lake over the last 15 years. Dissolved organic matter concentrations were moderate and above for most reservoirs (Figure 18). Nitrate values ranged widely. The Maryland reservoirs examined ranged from meso/oligotrophic to eutrophic, and this is reflected in NO_3 levels. However, sulfate levels varied by less than a factor of three across all systems. All but four of the reservoirs contained low-O_2 (<1 ppm) or anoxic bottom waters (Figure 18). In general, the western reservoirs were more dilute, contained lower levels of nutrients, particulate C, N, and DOC, and were less turbid. However, sulfate concentrations were generally higher in western reservoirs (Figure 19). Sulfate levels were generally lower in bottom waters, indicating depletion due to sulfate reduction. However, very little sulfide accumulated in bottom waters of any of the systems (Figure 19).

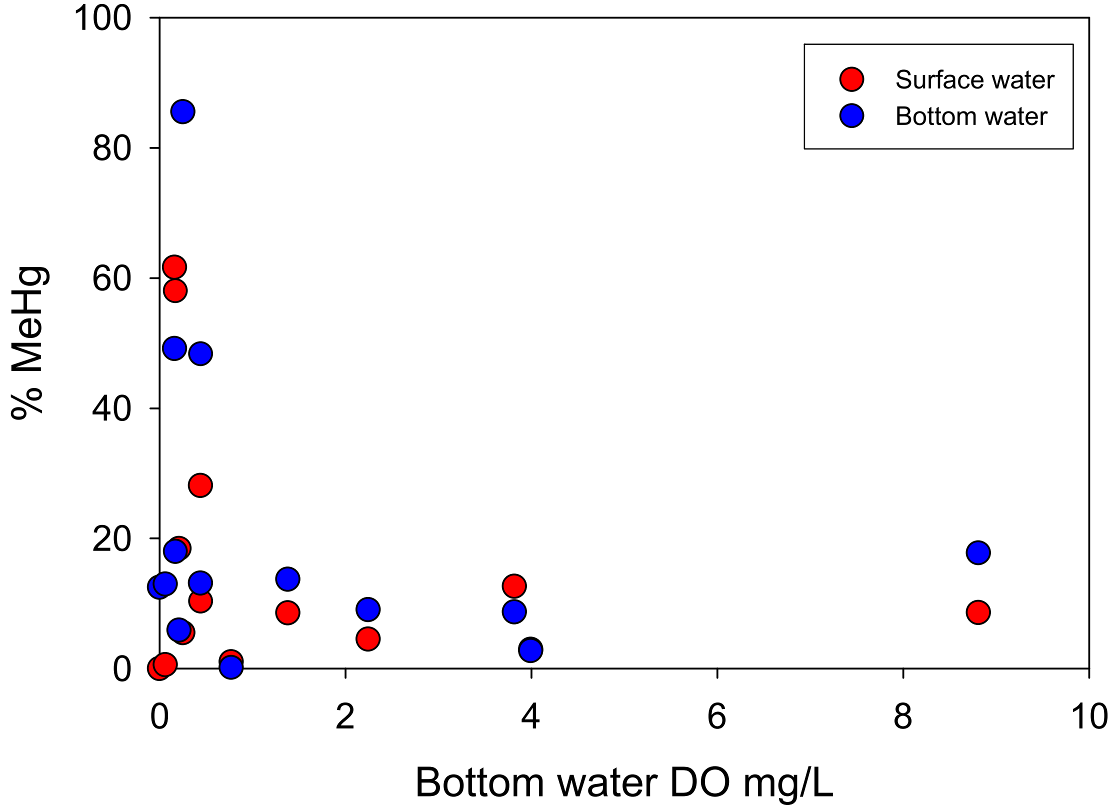
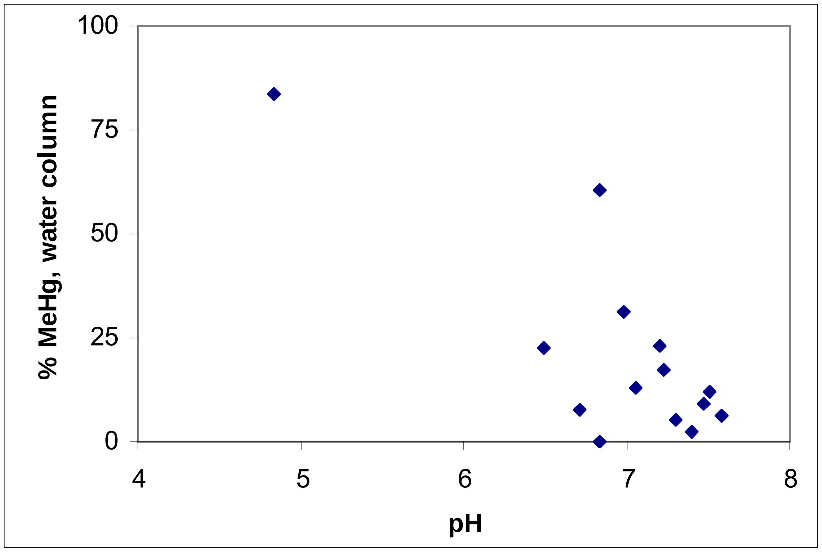


Figure 15. The relationship between bottom water dissolved oxygen and % MeHg for MD reservoirs, 2003-2005. Red is surface water %MeHg; blue is bottom water %MeHg, both are plotted against bottom water DO.



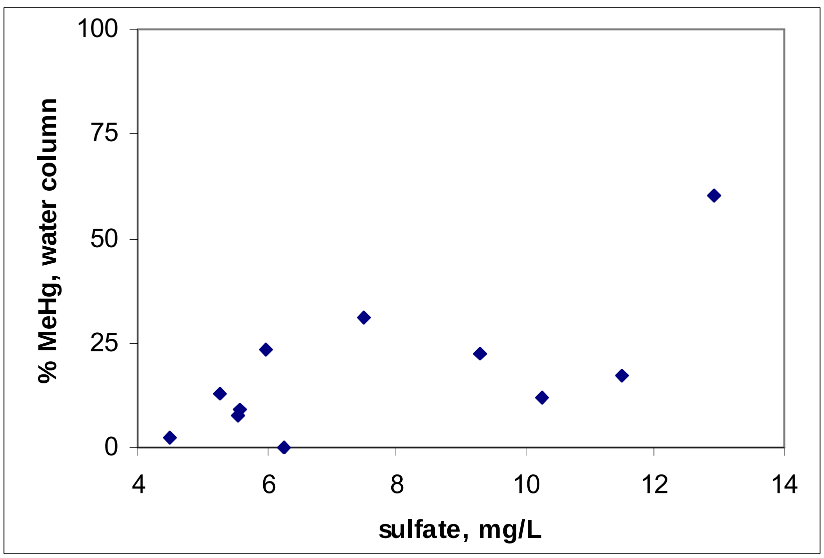


Figure 16. Significant relationships between %MeHg ($MeHg_D/Hg_D$) in reservoir surface waters and water chemistry variables. Data points are the average values of each variable for 2000-2005. Note that correlations were examined using log transformed variables, in order to normalize data distributions. Statistics for all correlations are in Appendix 1.

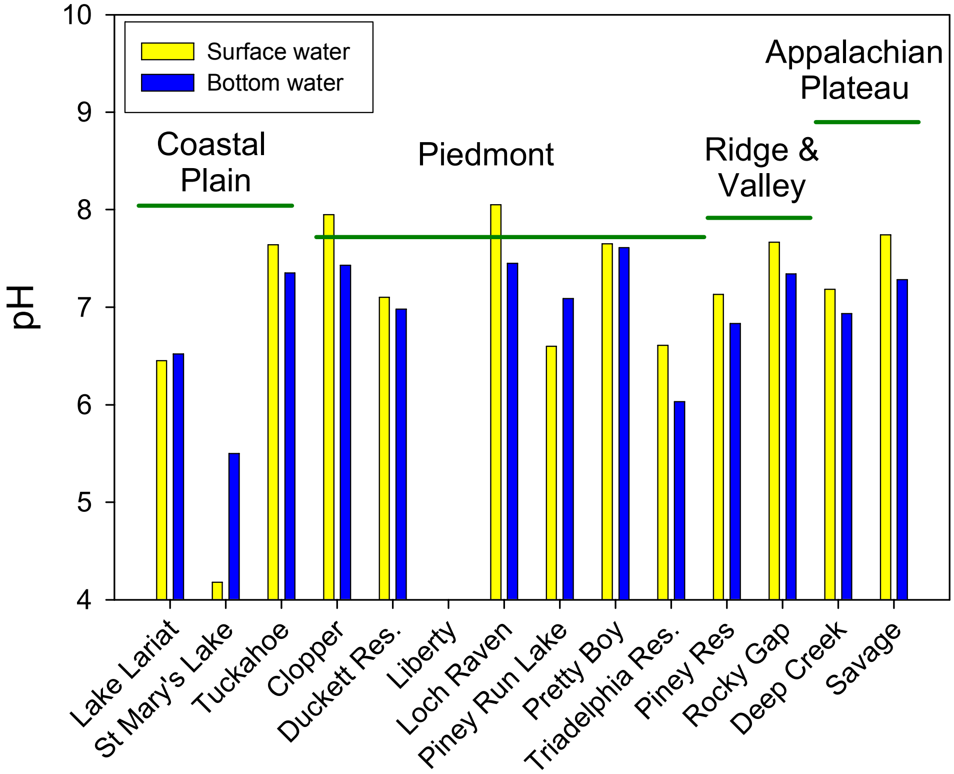


Figure 17. Reservoir pH values, 2003-2005. Data were not collected for Liberty Reservoir.

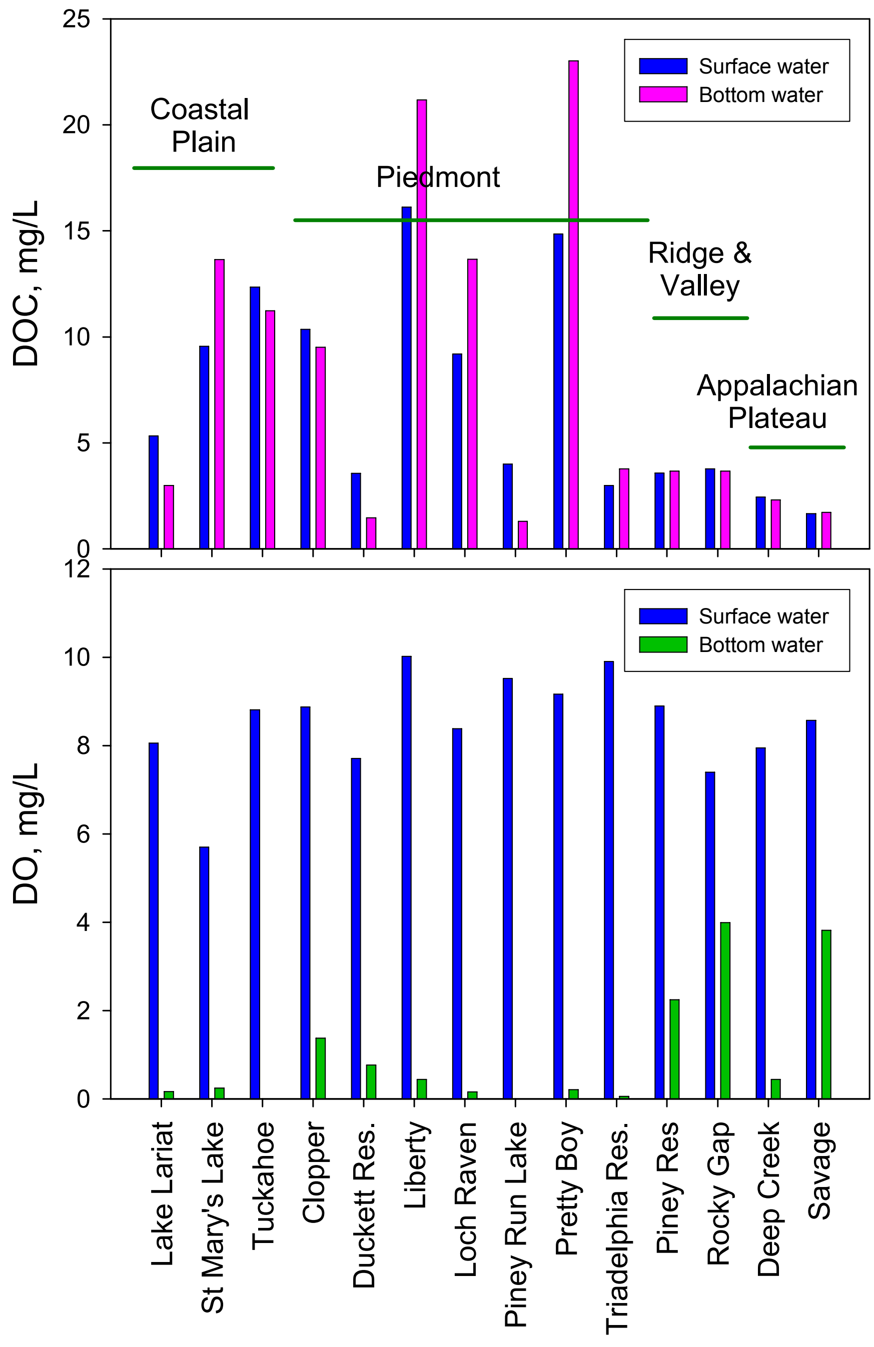


Figure 18. Reservoir dissolved organic carbon (top) and dissolved oxygen (bottom) concentrations, 2003-2005.

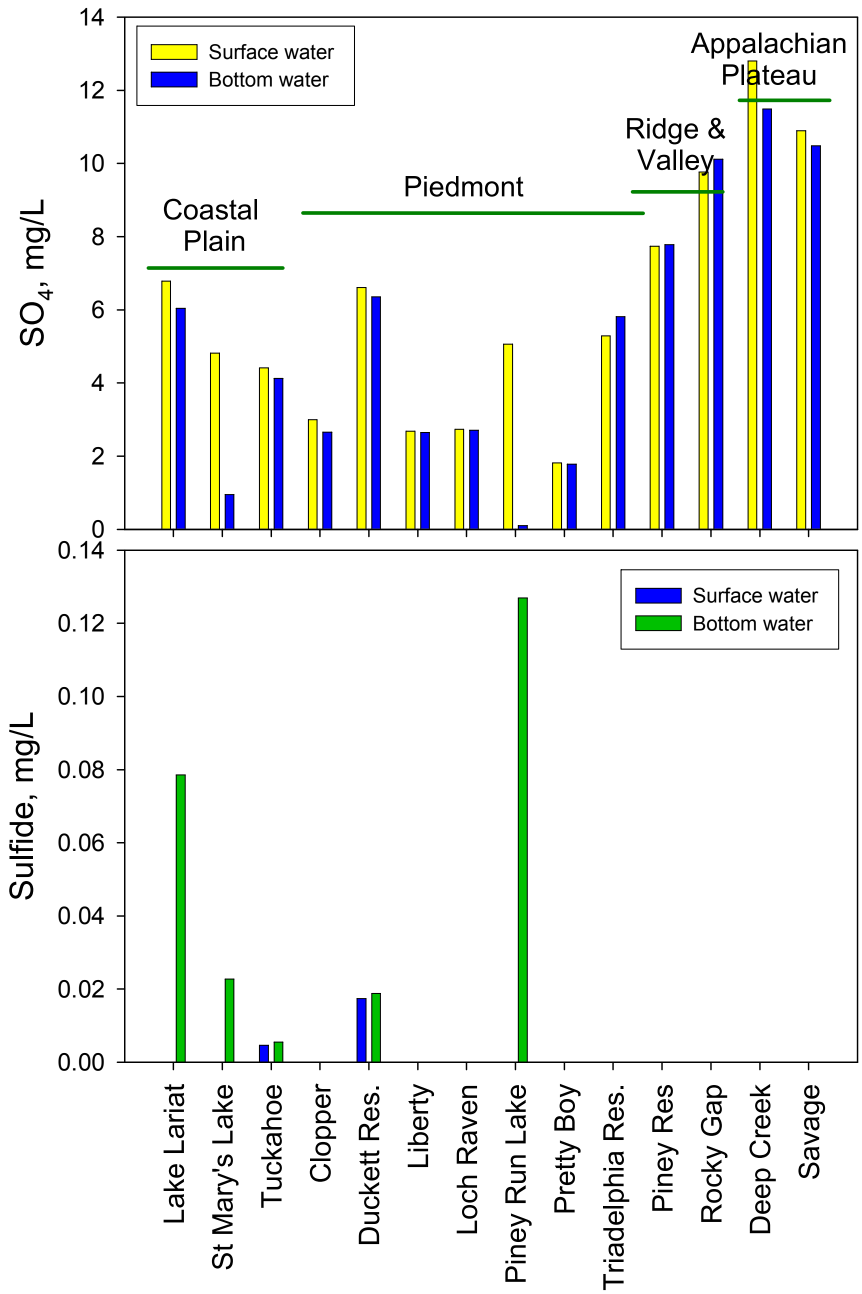


Figure 19. Reservoir sulfate (top) and dissolved sulfide (bottom) concentrations, 2003-2005. Sulfide was not measured in the western reservoirs; for eastern reservoirs without data, lake sulfide was below the method detection limit (BDL).

Sediment chemistry. For the eastern 10 reservoirs, sediments were sampled at three sites (S1 - shallow; S2 - intermediate depth; D – deepest part of the lake) in each of the 10 eastern reservoirs once during 2003-2004. Sediment bulk phase and pore water Hg and MeHg concentrations for the eastern reservoirs are given in Figures 20 and 21 and Tables 13-15, along with calculated sediment:pore water partition coefficients (K_D). Western reservoir sediments were sampled between 2003 and 2005. Up to five sites per reservoir were sampled, and most sites were sampled repeatedly across 2-3 years. Table 13b gives the depth of each site, the average bulk Hg and MeHg concentrations, and loss on ignition for the western reservoir sediments.

In many reservoirs, mercury concentrations were higher in deeper sediments (Figure 21), because Hg is generally correlated with the organic matter content of sediments (Figure 22A), which are often highest in the deepest, most focused sediment. Total mercury concentrations, expressed on a dry weight basis, were similar among most of the lakes. However, concentrations in Loch Raven Reservoir were roughly double the average values among the lakes. Dry weight Hg concentrations were highly correlated with all measures of sediment density, grain size or organic matter content.

Sediment MeHg concentrations varied somewhat more among the lakes, as did the depth of maximum MeHg concentration. Methylmercury concentrations in sediments were strongly related to total Hg concentrations (Figure 22B) and to variables that co-correlate with organic matter concentration. Sediment chemistry variables most related to % MeHg – used as a surrogate for net MeHg production, and to normalize for Hg content of sediments – were the reduced S content of sediments, the organic content of sediments, and the concentration of Fe(II) in sediment pore waters. Both reduced sulfur and reduced Fe in porewaters were positively related to MeHg in sediments (and to organic matter content), but negatively related to %MeHg. Methylmercury production is sensitive to the concentration of reduced sulfur compounds; these can in turn be affected by reduced iron concentrations.

Pore water chemistry data are shown in Table 13 and 14 and Figure 23. Bulk phase chemistry is given in Table 15, and in Figures 24 and 25. Sediments ranged widely in bulk density and from somewhat organic to highly organic. Coastal Plain reservoirs St. Mary’s and Lake Lariat had high pore water iron levels, reflecting an excess of reduced Fe over reduced sulfur, and high concentrations of reduced FeS minerals (acid-volatile sulfides - AVS and chromium reducible sulfides -CRS).

Table 13a. Sediment Hg and MeHg concentrations in the 10 eastern reservoirs sampled 2003-2004 by SERC/CBL, including solid phase and pore water concentrations, %MeHg (MeHg/Hg*100) for bulk and pore water phases, and partition coefficients, where K_D = (ng Hg/kg)/(ng Hg/L). BDL = below detection limit.

Sampling Date	Lake	Site	Water depth (m)	Hg (ng/L)	MeHg (ng/L)	%MeHg	HgT ng/gdw	MeHg ng/gdw	%MeHg	K_D Hg	K_D MeHg
7/15/2004	Clopper	S1	1.3	0.65	0.03	5.1	71.4	0.19	0.27	1.09E+05	5.82E+03
7/15/2004	Clopper	S2	3.9	1.90	0.09	4.6	94.4	0.43	0.46	4.96E+04	4.91E+03
7/15/2004	Clopper	D1	7.3	2.35	0.59	25.0	133.8	1.99	1.49	5.68E+04	3.38E+03
8/27/2003	Duckett	S1	0.6	1.53	0.17	11.4	22.9	0.11	0.47	1.49E+04	6.19E+02
8/27/2003	Duckett	S2	3.9	1.89	0.17	9.1	54.8	0.53	0.97	2.90E+04	3.09E+03
8/27/2003	Duckett	D1	19	2.51	0.18	7.1	97.5	0.65	0.67	3.89E+04	3.66E+03
7/29/2003	Lariat	S1	1.5	1.40	0.53	37.6	86.2	1.63	1.89	6.14E+04	3.09E+03
7/29/2003	Lariat	S2	3.9	0.87	0.28	31.8	106.3	1.01	0.95	1.22E+05	3.64E+03
7/29/2003	Lariat	D1	8.1	1.80	0.06	3.5	92.8	0.62	0.67	5.16E+04	1.00E+04
7/21/2004	Liberty	S1	0.9	2.13	1.37	64.4	52.9	1.05	1.98	2.49E+04	7.64E+02
7/21/2004	Liberty	S2	1.4	0.98	0.41	42.3	87.9	0.87	0.99	8.98E+04	2.10E+03
7/21/2004	Liberty	D1	15.2	2.62	1.00	37.9	121.6	1.30	1.07	4.64E+04	1.30E+03
9/22/2004	Loch Raven	S1	0.9	1.39	0.13	9.3	115.5	1.43	1.24	8.28E+04	1.11E+04
9/22/2004	Loch Raven	S2	3.5	0.43	0.02	4.9	187.0	0.37	0.20	4.37E+05	1.75E+04
9/22/2004	Loch Raven	D1	16.7	0.83	0.15	18.2	325.6	3.05	0.94	3.91E+05	2.01E+04
8/29/2003	Piney Run	S1	1.1	1.21	BDL		38.8	0.22	0.56	3.20E+04	
8/29/2003	Piney Run	S2	4.2	1.04	0.01	1.1	67.1	0.15	0.23	6.47E+04	1.40E+04
8/29/2003	Piney Run	D1	14.7	0.62	0.08	12.5	81.9	0.78	0.95	1.31E+05	9.95E+03
9/24/2004	Pretty Boy	S1	1.2	1.44	0.30	20.6	38.5	0.80	2.07	2.68E+04	2.70E+03
9/24/2004	Pretty Boy	S2	4.6	4.00	0.92	23.0	59.3	0.97	1.64	1.48E+04	1.06E+03
9/24/2004	Pretty Boy	D1	13.7	1.15	0.35	30.1	122.4	0.36	0.30	1.06E+05	1.04E+03
7/31/2003	St. Mary's	S1	1.6	2.85	0.29	10.2	25.6	0.26	1.01	8.96E+03	8.91E+02
7/31/2003	St. Mary's	S2	3.9	3.67	0.38	10.4	55.8	0.40	0.73	1.52E+04	1.06E+03
7/31/2003	St. Mary's	D1	6.5	4.67	0.34	7.2	77.9	0.59	0.75	1.67E+04	1.74E+03
8/25/2003	Triadelphia	S1	1.3	1.89	0.31	16.6	24.8	0.17	0.69	1.31E+04	5.41E+02
8/25/2003	Triadelphia	S2	3.9	1.61	0.27	16.5	44.9	0.67	1.49	2.79E+04	2.52E+03
8/25/2003	Triadelphia	D1	13.5	3.49	0.87	25.1	69.2	0.78	1.13	1.98E+04	8.95E+02
10/6/2004	Tuckahoe	S1	<1	1.02	0.11	10.9	142.4	0.73	0.51	1.40E+05	6.55E+03
10/6/2004	Tuckahoe	S2	0.6	0.43	0.04	9.8	130.3	1.39	1.07	3.00E+05	3.29E+04
10/6/2004	Tuckahoe	D1	1.2	0.97	0.17	17.0	13.5	0.14	1.01	1.39E+04	8.26E+02

Table 13b. Sediment Hg and MeHg concentrations in the 4 western reservoirs sampled 2003-2005 by AL.

Lake	Water depth (m)	LOI %	HgT ng/gdw	MeHg ng/gdw	%MeHg	n
Piney Reservoir	2.0	10.0	45.6	0.57	1.16	5
Piney Reservoir	5.0	8.6	68.4	0.50	0.85	5
Piney Reservoir	9.0	10.6	83.2	0.63	0.83	5
Rocky Gap	2.6	10.7	50.2	0.19	0.58	4
Rocky Gap	7.2	8.7	39.7	0.16	0.56	4
Rocky Gap	8.8	8.9	36.0	0.16	0.56	4
Rocky Gap	9.7	7.9	23.0	0.16	0.69	4
Rocky Gap	9.9	3.9	20.3	0.10	1.03	4
Savage Reservoir	2.5	10.8	48.6	2.31	4.79	3
Savage Reservoir	7.0	10.9	45.6	1.45	3.17	4
Savage Reservoir	11.0	7.1	36.4	0.70	1.91	4
Savage Reservoir	12.5	9.5	44.3	1.04	2.38	4
Savage Reservoir	12.8	3.4	53.2	0.48	0.80	2
Deep Creek Lake	8.7	9.4	68.3	0.19	0.27	4
Deep Creek Lake	9.7	6.6	43.6	0.20	0.36	4
Deep Creek Lake	11.3	12.7	53.4	0.83	1.91	4
Deep Creek Lake	13.2	10.4	79.3	0.69	0.88	4
Deep Creek Lake	15.0	7.5	75.6	0.31	0.38	4

Table 14. Sediment pore water chemistry for 10 eastern reservoirs, 2003-2004. Iron values in red exceed the range of standards and are estimates.

Sampling Date	Lake	Site	Water depth (m)	pH	Sulfide (mg/L)	Fe (ug/L)	Mn (ug/L)	F (mg/L)	Cl (mg/L)	NO3 (mg/L)	PO4 (mg/L)	SO4 (mg/L)	DOC (mg/L)
7/15/2004	Clopper	S1	1.3	7.11	0.45	10.9	0.03	0.01	76.5	0.00	BDL	0.17	14.0
7/15/2004	Clopper	S2	3.9	7.15	0.31	17.3	0.04	0.13	140.0	-0.01	BDL	0.51	6.6
7/15/2004	Clopper	D1	7.3	2.59	11.52	21.9	0.03	0.25	511.9	0.02	0.80	0.47	9.3
8/27/2003	Duckett	S1	0.6	No data	0.03	22	3.92	0.06	22.2	0.22	BDL	1.07	6.64
8/27/2003	Duckett	S2	3.9	No data	0.03	34	5.19	0.07	18.6	0.47	BDL	0.35	22.98
8/27/2003	Duckett	D1	19	No data	0.02	30	21.20	0.40	22.8	0.57	BDL	0.53	31.29
7/29/2003	Lariat	S1	1.5	No data	0.01	23	0.94	0.01	19.4	0.67	BDL	0.63	11.74
7/29/2003	Lariat	S2	3.9	No data	BDL	30	1.61	0.08	20.0	0.40	BDL	-0.04	13.13
7/29/2003	Lariat	D1	8.1	No data	BDL	~2500	1.82	0.11	22.7	1.96	0.14	0.59	24.46
7/21/2004	Liberty	S1	0.9	No data	No data	47.4	0.13	0.10	193.7	0.19	BDL	0.87	17.6
7/21/2004	Liberty	S2	1.4	No data	No data	3.4	0.07	0.23	105.5	0.07	BDL	0.84	14.3
7/21/2004	Liberty	D1	15.2	No data	No data	60.1	0.13	0.21	110.8	0.02	BDL	0.20	21.0
9/22/2004	Loch Raven	S1	0.9	7.57	BDL	1.2	0.06	0.28	53.0	-0.01	BDL	5.54	10.0
9/22/2004	Loch Raven	S2	3.5	7.44	BDL	9.2	0.03	0.13	54.3	-0.01	BDL	0.36	8.2
9/22/2004	Loch Raven	D1	16.7	7.12	BDL	32.8	0.11	0.52	319.2	0.03	BDL	0.94	13.7
8/29/2003	Piney Run	S1	1.1	No data	BDL	15	1.66	0.08	21.8	0.39	BDL	0.80	12.14
8/29/2003	Piney Run	S2	4.2	No data	BDL	11	5.41	0.06	21.1	0.34	BDL	0.37	16.88
8/29/2003	Piney Run	D1	14.7	No data	BDL	34	5.25	0.08	24.5	0.74	BDL	1.04	23.73
9/24/2004	Pretty Boy	S1	1.2	No data	0.28	25.5	0.10	-0.02	14.8	5.48	BDL	0.47	15.6
9/24/2004	Pretty Boy	S2	4.6	No data	0.30	2.1	0.11	0.41	58.9	0.08	BDL	0.32	13.9
9/24/2004	Pretty Boy	D1	13.7	No data	BDL	55.9	0.09	0.27	34.2	0.02	BDL	0.03	23.0
7/31/2003	St. Mary's	S1	1.6	No data	0.01	2263	3.24	0.17	7.0	1.08	0.17	0.67	28.45
7/31/2003	St. Mary's	S2	3.9	No data	0.01	379	0.94	0.14	8.2	1.03	0.22	0.68	17.35
7/31/2003	St. Mary's	D1	6.5	No data	0.01	~2500	1.14	2.64	6.9	3.39	0.54	3.00	32.6
8/25/2003	Triadelphia	S1	1.3	No data	BDL	6	1.42	0.04	21.5	0.08	0.46	1.52	6.92
8/25/2003	Triadelphia	S2	3.9	No data	BDL	14	3.37	0.05	25.0	0.69	BDL	2.05	20.16
8/25/2003	Triadelphia	D1	13.5	No data	BDL	27	7.70	0.07	29.0	1.00	BDL	1.24	23.53
10/6/2004	Tuckahoe	S1	<1	7.36	BDL	22.6	0.04	0.25	12.3	0.17	BDL	3.02	9.0
10/6/2004	Tuckahoe	S2	0.6	7.10	BDL	7.7	0.02	0.17	9.6	0.29	0.04	2.25	13.3
10/6/2004	Tuckahoe	D1	1.2	7.28	BDL	7.3	0.02	0.21	12.0	0.34	0.05	2.60	12.3

Table 15. Sediment bulk phase chemistry for 10 eastern reservoirs, 2003-2004.

Sampling Date	Lake	Site	Water depth (m)	Bulk density g/cm3	Dry Wt g/cm3	Porosity ml/cm3	LOI %	AVS umoles/g dw	CRS umoles/g dw	Extractable Fe(II) mg/gdw	Extractable Fe(III) mg/gdw
7/15/2004	Clopper	S1	1.3	1.15	0.30	0.74	10.8	7.74	22.2	9.0	BDL
7/15/2004	Clopper	S2	3.9	1.23	0.37	0.70	1.2	2.60	13.4	11.2	BDL
7/15/2004	Clopper	D1	7.3	1.09	0.16	0.85	13.9	72.81	78.1	54.9	BDL
8/27/2003	Duckett	S1	0.6	1.58	0.99	0.37	2.4	1.54	9.8	4.2	BDL
8/27/2003	Duckett	S2	3.9	1.25	0.49	0.61	6.3	0.92	15.1	6.7	1.15
8/27/2003	Duckett	D1	19	1.06	0.18	0.83	10.6	6.40	14.3	37.9	BDL
7/29/2003	Lariat	S1	1.5	1.12	0.26	0.77	12.9	4.20	22.6	13.0	BDL
7/29/2003	Lariat	S2	3.9	1.04	0.19	0.81	11.6	55.67	109.1	15.5	BDL
7/29/2003	Lariat	D1	8.1	1.09	0.21	0.81	12.8	516.97	452.6	34.2	0.40
7/21/2004	Liberty	S1	0.9	1.26	0.45	0.64	9.7	0.42	3.8	13.7	BDL
7/21/2004	Liberty	S2	1.4	1.35	0.51	0.62	8.7	0.37	4.6	7.6	BDL
7/21/2004	Liberty	D1	15.2	1.15	0.22	0.81	11.8	6.80	19.6	15.3	BDL
9/22/2004	Loch Raven	S1	0.9	1.26	0.44	0.65	12.6	0.21	5.1	7.7	0.17
9/22/2004	Loch Raven	S2	3.5	1.09	0.29	0.73	10.8	8.19	22.8	12.2	BDL
9/22/2004	Loch Raven	D1	16.7	0.97	0.04	0.96	20.6	204.31	293.3	115.8	BDL
8/29/2003	Piney Run	S1	1.1	1.30	0.55	0.58	5.3	11.50	45.7	5.7	BDL
8/29/2003	Piney Run	S2	4.2	1.01	0.23	0.78	8.1	5.43	23.5	27.7	BDL
8/29/2003	Piney Run	D1	14.7	1.04	0.15	0.86	11.2	18.16	51.5	40.9	1.19
9/24/2004	Pretty Boy	S1	1.2	1.33	0.59	0.56	7.4	0.20	2.2	0.9	0.68
9/24/2004	Pretty Boy	S2	4.6	1.27	0.46	0.64	9.5	0.15	2.6	6.3	BDL
9/24/2004	Pretty Boy	D1	13.7	1.17	0.26	0.78	10.2	1.05	57.3	22.8	BDL
7/31/2003	St. Mary's	S1	1.6	1.51	0.93	0.38	2.6	0.42	3.5	3.4	5.27
7/31/2003	St. Mary's	S2	3.9	1.22	0.47	0.62	6.5	5.99	62.9	9.2	0.05
7/31/2003	St. Mary's	D1	6.5	1.16	0.35	0.70	6.4	34.65	169.3	16.5	BDL
8/25/2003	Triadelphia	S1	1.3	1.42	0.76	0.47	4.9	1.72	10.5	2.9	BDL
8/25/2003	Triadelphia	S2	3.9	1.27	0.47	0.63	7.1	0.75	7.5	6.3	BDL
8/25/2003	Triadelphia	D1	13.5	1.11	0.21	0.81	8.6	7.70	18.7	18.4	0.47
10/6/2004	Tuckahoe	S1	<1	1.14	0.31	0.73	17.2	3.85	24.1	7.8	BDL
10/6/2004	Tuckahoe	S2	0.6	1.02	0.13	0.87	29.7	2.40	34.6	13.9	0.06
10/6/2004	Tuckahoe	D1	1.2	1.41	0.61	0.57	6.9	0.86	7.7	2.0	0.29

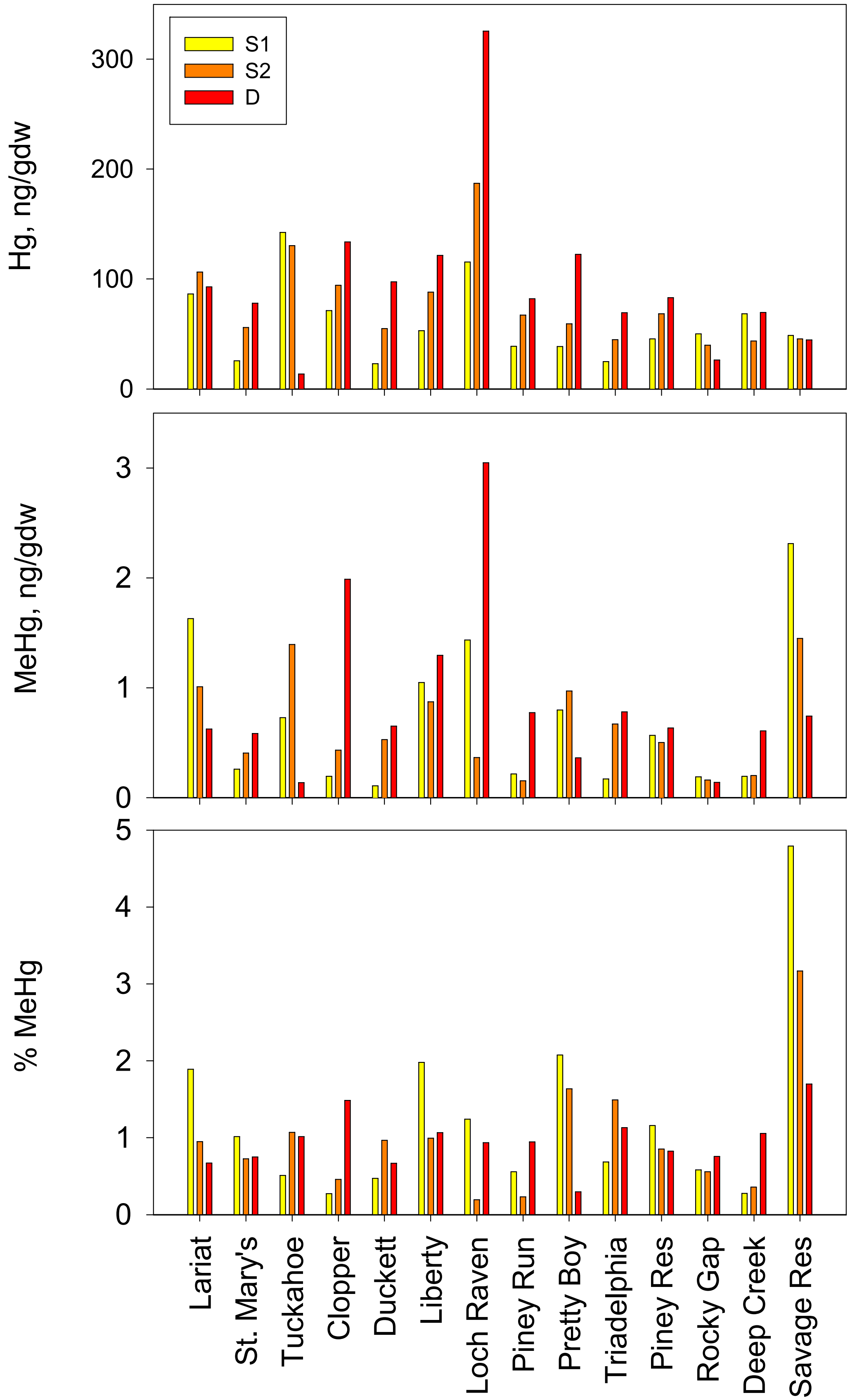


Figure 20. Sediment bulk phase Hg and MeHg concentrations, and %MeHg (MeHg/Hg*100), for 3 sites in each reservoir. Site S1 sediments were sampled at <3 m water depth; site S2 sediments were sampled at <5 m water depth; and site D were sampled at the maximum water depth. Water sampling depths are given in Table 13. All data for top 4 cm of sediments. Data from eastern reservoirs are for one date in 2003 or 2004; data for the western reservoirs are composites for multiple dates in 2003-2005.

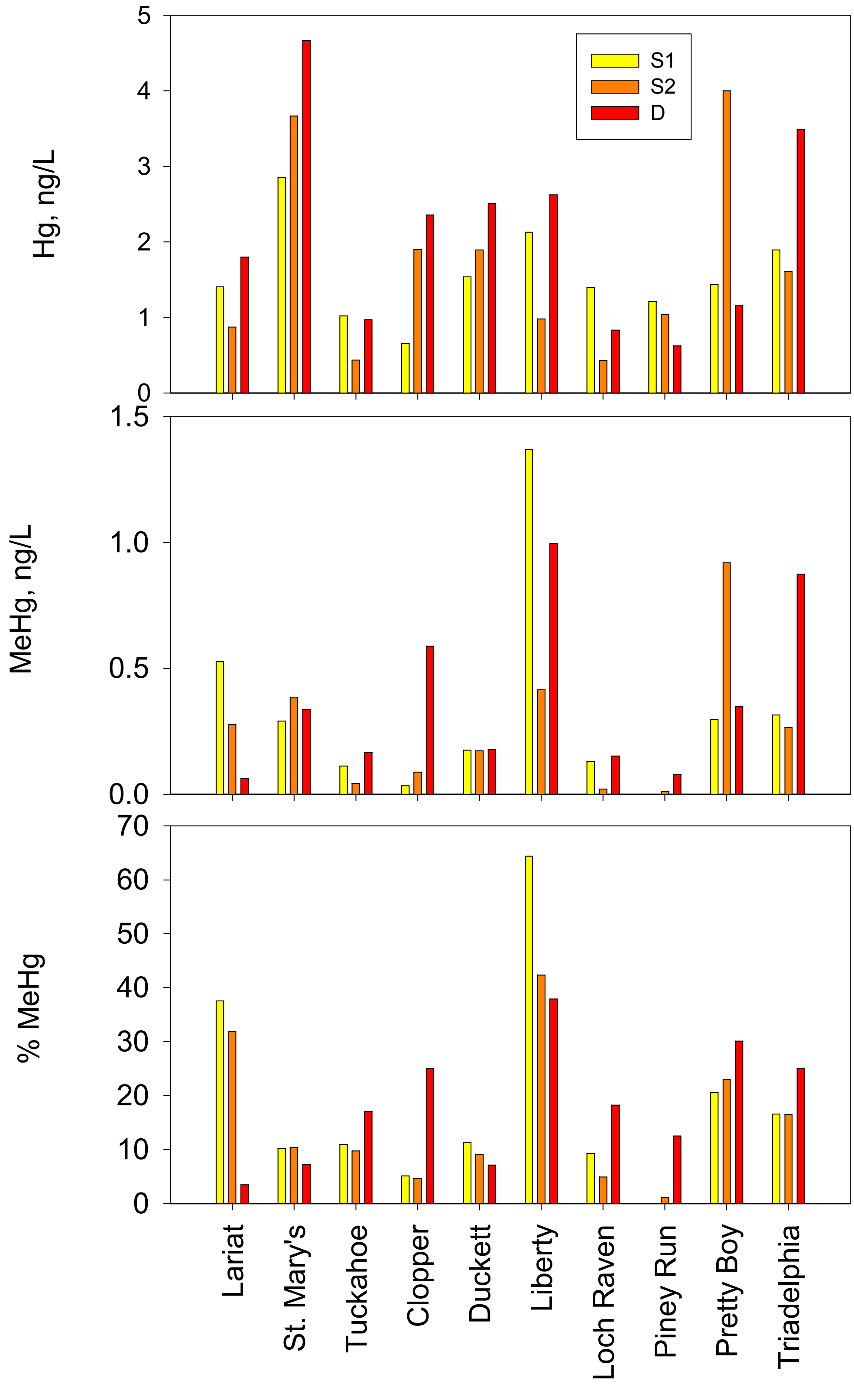


Figure 21. Sediment pore water total Hg and MeHg concentrations, and %MeHg for 3 sites in each reservoir. Site S1 sediments were sampled at <3 m water depth; site S2 sediments were sampled at <5 m water depth; and site D were sampled at the maximum water depth. Water sampling depths are given in Table 13. All data are for the top 4 cm of sediments.

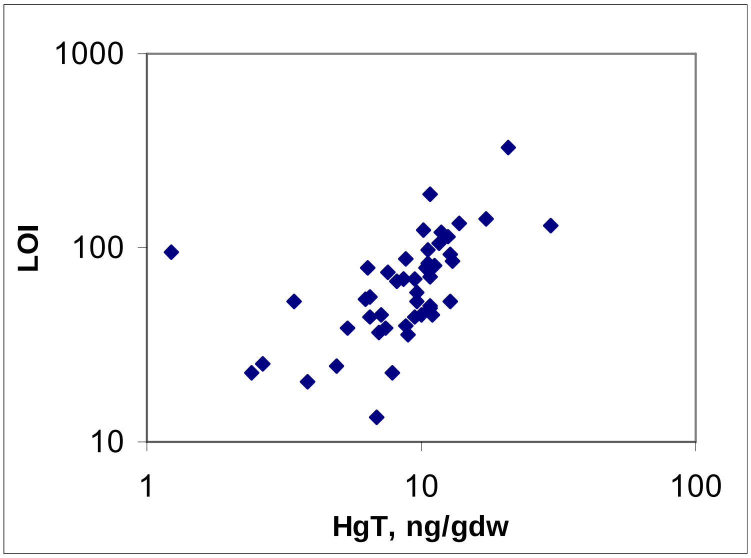


Figure 22A. The relationship between organic matter content of sediments (as measured by loss-on-igntition) and sediment Hg concentration ($r^2 = 0.47$; $P < 0.006$ for log transformed variables). Sediment Hg content generally increases with loss-on-ignition (LOI) because of the affinity of Hg for organic matter.

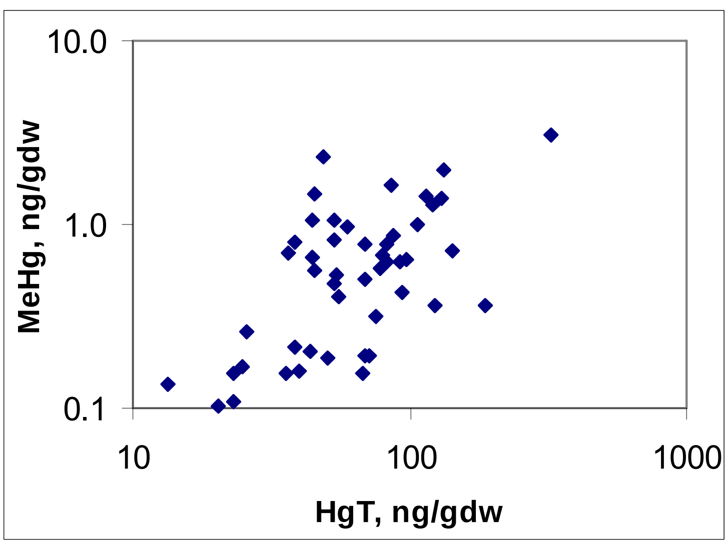


Figure 22B. The relationship between total Hg and MeHg concentrations in sediments ($r^2 = 0.54$; $P < 0.003$). Note that correlations were examined using log transformed variables, in order to normalize data distributions. Statistics for all correlations are in Appendix 1.

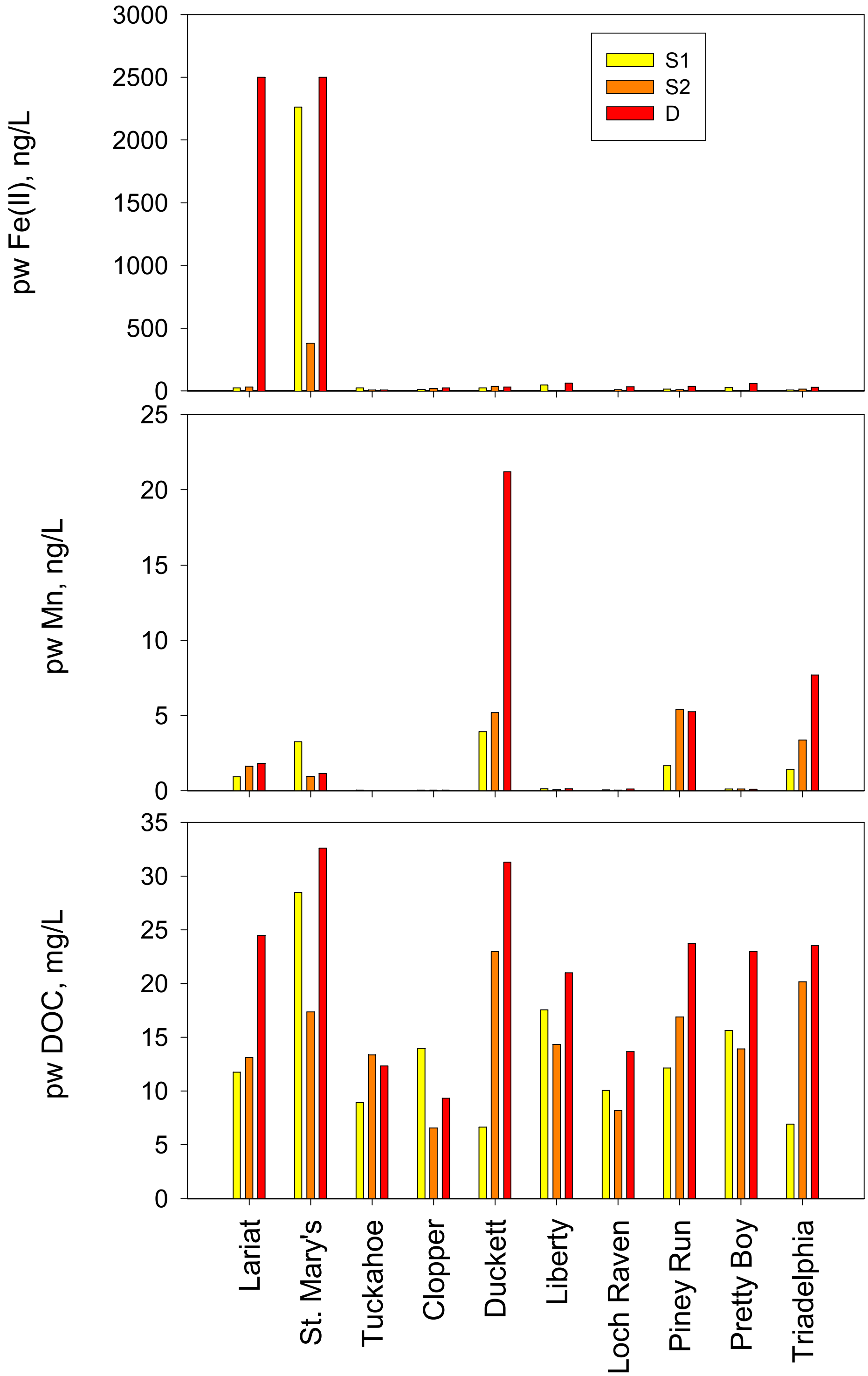


Figure 23. Sediment pore water Fe, Mn and DOC concentrations for 3 sites in each reservoir. Site S1 sediments were sampled at <3 m water depth; site S2 sediments were sampled at <5 m water depth; and site D were sampled at the maximum water depth. Water sampling depths are given in Table 13. All data are for top 4 cm of sediments.

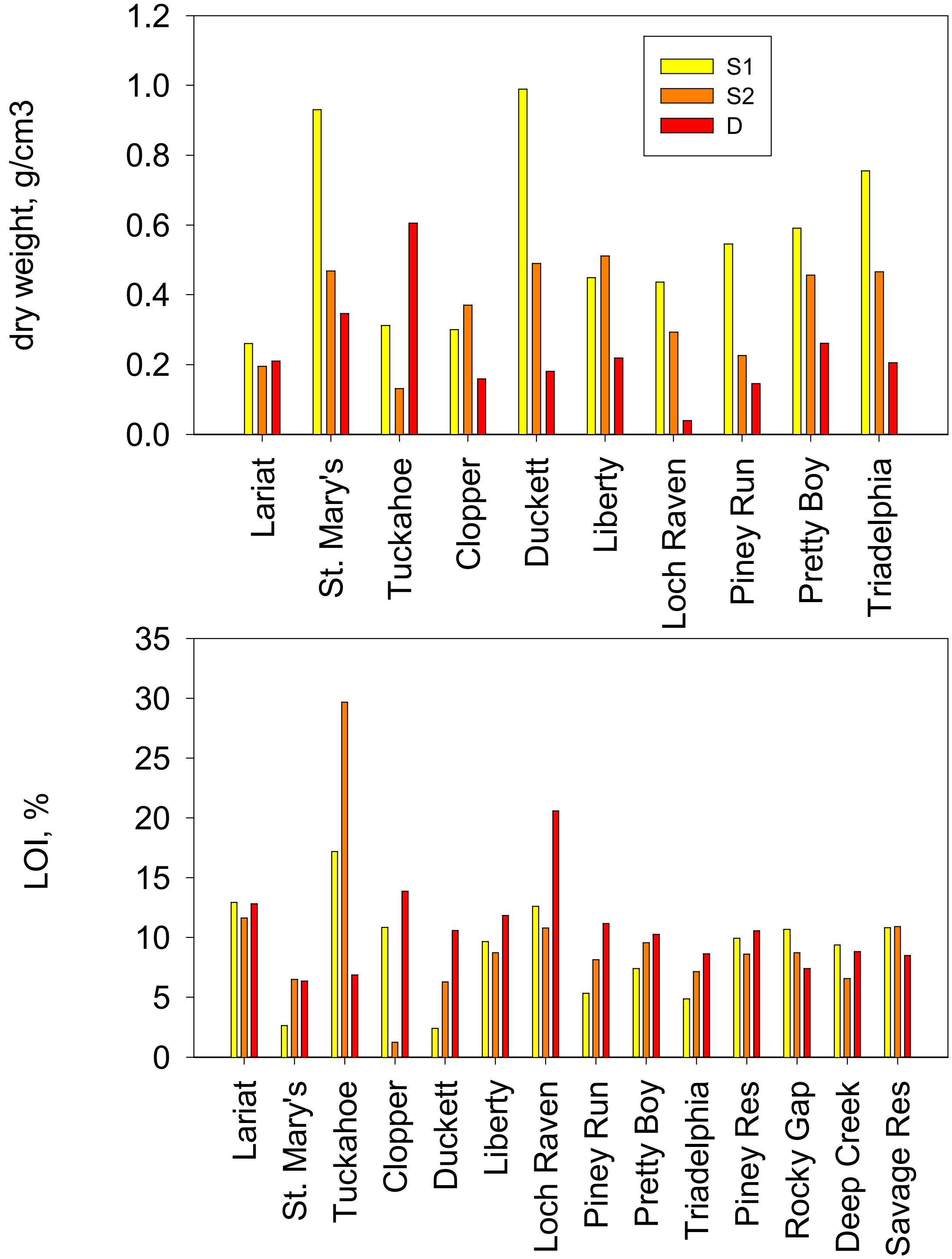


Figure 24. Sediment dry weight and loss on ignition (LOI) for 3 sites in each reservoir. Site S1 sediments were sampled at <3 m water depth; site S2 sediments were sampled at <5 m water depth; and site D were sampled at the maximum water depth. Water sampling depths are given in Table 13. All data are for the top 4 cm of sediments.

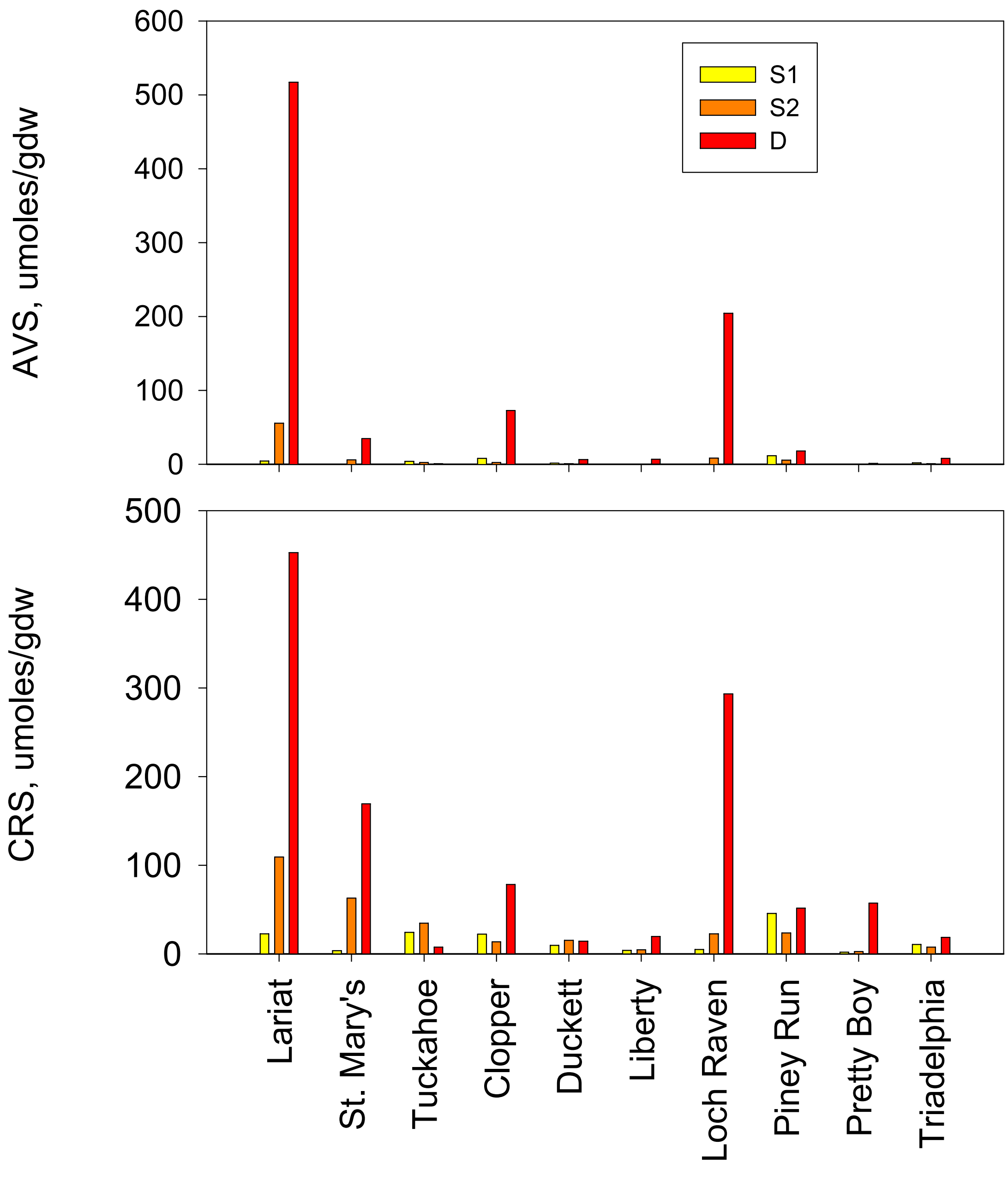


Figure 25. Reduced sulfur content of sediments, AVS (top) and CRS (bottom). Site S1 sediments were sampled at <3 m water depth; site S2 sediments were sampled at <5 m water depth; and site D were sampled at the maximum water depth. Water sampling depths are given in Table 13. All data are for the top 4 cm of sediments.

Statistical Analysis

Each of the major components of the Hg cycle was modeled separately, and then an overall model for Hg in largemouth bass was examined. Although we wish to understand the parameters that control MeHg concentrations in fish, a regression analysis of fish Hg concentrations against all the other measured variables could obscure other important relationships which impact Hg transport, methylation and bioaccumulation. The component processes that were modeled are shown below. For each component, a key dependent variable or variables was chosen. For example, to examine the factors that relate to Hg accumulation in lakes, Hg in sediments and water were examined as dependent variables.

Component Process in Hg cycle	Dependent variables assessed
Hg deposition, transport and accumulation in lakes	Hg in water and sediments
Net MeHg production	MeHg in water and sediments
MeHg bioaccumulation	Bioaccumulation factors for size-normalized largemouth bass
Overall	Size-normalized Hg in largemouth bass

Statistical methods. Single and stepwise multiple regression models were used to assess relationships using SAS release 8.02. Prior to regression analysis, all parameters were tested for normal frequency distribution, and were transformed as needed prior to further analysis. Natural log transformations were used for many variables. All parameters could be transformed to meet normalcy requirements except sulfide and manganese in sediment pore waters, and extractable Fe(III) in sediments. These variables were excluded from further analyses.

After the data were checked for normality and transformed as needed, a correlation matrix for all variables was constructed (Appendix 1). Although the correlation matrix guided further analyses, models for each component of the Hg cycle were constructed using stepwise linear regression. This approach identifies variables in order of significance, and more importantly, allows correlations with residual variability to be assessed. This approach provides information that a simple correlation matrix of all individual variables does not. However, it can be compromised by co-correlations within the model.

Factors related to Hg deposition, transport and accumulation in lakes. The concentrations of Hg in sediment and water were used as dependent variables to examine relationships with deposition and transport variables. However, prior to constructing models for deposition and transport, the variability in these parameters due to sediment and water chemistry was examined. These factors were then incorporated into models of watershed parameters.

Sediments: Relationships between Hg concentration and sediment physical and chemical properties. The data set for the 10 eastern reservoirs for which sediment chemistry data were available was used for this analysis. All 3 sites in each reservoir were included individually in the analysis, giving n =30. The final data set used for this analysis was taken from Tables 13-15.

Correlations between Hg in sediment and individual sediment chemistry variables were significant for all measures related to sediment grain size and organic matter content. Parameters related to redox and the reduced sulfur content were also strongly related to total Hg concentrations in sediments. Mercury strongly sorbs organic matter and reduced sulfur components in sediments, so measures of sediment grain size, organic matter content, and reduced sulfur components were expected to be highly correlated with Hg in sediments. Sediment Hg content increased with water depth in many reservoirs (Figure 21). This relationship influenced the sediment sampling design, which called for samples from 3 different water depths in each lake, from similar depth intervals where possible.

The stepwise linear regression model used to assess relationships between Hg in sediment and sediment physical and chemical properties is shown below. The model statement (taken directly from SAS) lists the variables used in the initial stepwise regression. Only the variables that were significant were retained; these variables listed in each table as "variables included." Variable acronyms can be found in Appendix 1. The table shows the degrees of freedom for the model (generally the number of observations used in the model minus 1); the p value for the entire model; the variables included in the model, their partial r^2 within the model, and p for each variable. The p to enter was set < 0.15.

Model for sediment Hg (sediment chemistry only)		
Model logHg_nggdw = DOC_mgL logwat_m bulkden por logdw logLOI logAVS logCRS logFeII log2FeugL/selection = stepwise		
DF	28	
Model r^2	0.68	
Model p	<0.0001	
Variables included	***Partial r^2***	***p***
por	0.68	<0.0001

This analysis showed that almost 70% of the variability in sediment Hg among the 30 sites examined could be explained by sediment porosity, which is essentially one measure of grain size. For further analysis of potential controls on Hg in sediments, sediment porosity or organic matter content (measured as LOI) were included in the stepwise analyses to normalize for this relationship. Sediment organic matter content was available for all 14 reservoirs.

Sediments: Relationships between sediment Hg concentration and watershed variables. To examine the relationships between Hg accumulation in sediments and watershed parameters, a stepwise linear regression model including the land use classes shown in Table 3, estimated total Hg deposition rates (Table 5; Garrison and Sherwell 2006), and the reservoir/watershed physical and hydrologic parameters in

Table 1 were used. The model also included sediment porosity to normalize for the effects of sediment chemistry.

For this analysis, the overall average values for each parameter in each reservoir were used (n=14). The final data set used for this analysis is given in Tables 16, 18 and 19. For water column Hg and MeHg data, the Svensdottir 2001-2002 data set was combined with the data collected in this study (see Table 17). In order to avoid bias from years when bottom water data were not collected, the values in Table 18 were calculated as the [(average of all surface water data) + (average of all bottom water data)]/2. The same method was used to calculate average water chemistry for other parameters. Parameters for which data were not available for many reservoirs were dropped from the overall analysis.

Model for sediment Hg		
model logsedHg = LOI age depth logsurfarea logcap logwatarea logsurftowat logflow restime HgdepDNR logHgD logHgUNF logLUdev logLUag logLUfor logLUwet/selection = stepwise		
DF	13	
Model r^2	0.85	
Model p	0.025	
Variables included	***Partial r^2***	***p***
LOI	0.48	0.001
logHgD	0.13	0.025
logLUdev	0.12	0.025
HgDepDNR	0.12	0.015

After removing the effects sediment organic matter content, these variables contributed significantly to the model: water column Hg concentrations, the percent of developed land, and the local Hg deposition rate.

Water: Relationships between water Hg concentrations and water chemistry. For this analysis, the average water column data set for all eastern and western reservoirs was used (Tables 16, 18 and 19), except Duckett reservoir, which was excluded from the analysis (n=13). The water column Hg data sets could not be normalized when the anomalously high Hg concentrations in Duckett were included. Filterable Hg was used as the dependent variable.

Model for water column HgD (water chemistry only)		
model logHgD = ph logTSS logDOC logCl logNO3 logSO4 logbottDO/selection = stepwise;		
DF	11	
Model r^2	0.818	
Model p	0.0005	
Variables included	***Partial r^2***	***p***
logCl	0.70	0.0007
DOC	0.12	0.04

To examine the relationships between water chemistry and filterable Hg concentrations in water, the model included all non-Hg water chemistry variables.
Only two variables contributed to the model, chloride and DOC. Filterable Hg concentrations increased with the concentrations of both.

Water: Relationships between water column Hg concentration and watershed variables. The stepwise model included land use, modeled total Hg deposition rates, reservoir/ watershed physical parameters, and surface water sulfate and DOC to normalize for the major effects of water chemistry on Hg concentrations among lakes. Although chloride showed the strongest correlation with Hg of the water chemistry variables examined, it was also strongly correlated with land use, while DOC was not.

Model for water column HgD		
model logHgD = age depth logsurfarea logsurftowat logflow restime logHgdep HgdepDNR logLUdev logLUag logLUfor logLUwet ph logTSS logDOC logSO4 logbottDO loi logsedHg logsedMeHg		
DF	12	
Model r^2	0.93	
Model p	0.0001	
Variables included	***Partial r^2***	***p***
logsedHg	0.44	0.014
logSO4	0.12	0.035
HgdepDNR	0.1	0.011
logsurfarea	0.02	0.12

Other than water chemistry, and Hg concentrations in sediments, the strongest correlate with filterable Hg concentrations in lakes were sulfate concentrations and local modeled Hg deposition rates. Both filterable and total Hg concentrations increased with the percent of developed land and decreased with the percent forest (Figure 26). The percent developed land was generally inversely related to the percent forested land.

Relationships between land use and water and sediment chemistry. Relationships between land use and water and sediment chemistry were also examined. Watersheds with higher percentages of developed land had significantly higher chloride concentrations, lower bottom water dissolved oxygen, and finer-grained sediments (Figure 27), all parameters predictive of either higher Hg or %MeHg in reservoirs.

Factors related to MeHg concentrations in water and sediments. The main locations of MeHg production in most reservoirs are likely to be sediments, and in some cases anoxic bottom waters. Wetlands in watershed may also produce MeHg which could be transported to the reservoirs, but transport of MeHg into these systems was not directly measured in this study. Methylmercury concentrations in sediments and bottom waters were used as endpoints to compare net MeHg production among the reservoirs.

Sediments. Total Hg concentrations in sediments account for about 55% of the variability in MeHg in sediments (Figure 22B). Therefore, Hg – the substrate for methylation – accounted for much but not all of the variability in MeHg concentration among these sites. MeHg concentrations in sediments were also correlated with most of the variables that predict Hg concentrations in sediments – organic matter, porosity, reduced Fe and S compounds. Although some studies have noted higher net MeHg production in shallow sediments, we found that the depth of maximum %MeHg was variable among the lakes.

MeHg concentrations in surface waters were also strongly related to bottom water anoxia, with much higher surface and bottom water %MeHg in stratified lakes with low or zero DO hypolimnia.

In order to assess the remainder of the variability in MeHg concentration, stepwise regression analysis was done using MeHg normalized to the total Hg content of sediment, here called %MeHg, where:

%MeHg = (ng MeHg/gdw sediment)/(ng HgT/gdw sediment) *100

Model for %MeHg in sediments (sediment chemistry)		
model perMeHgbulk = logHg_nggdw log2Fe_ugL DOC_mgL logwat_m logLOI logAVS logCRS/selection = stepwise;		
DF	28	
Model r^2	0.424	
Model p	0.0028	
Variables included	***Partial r^2***	***p***
logCRS	0.16	
logLOI	0.17	
log2Fe_ugL	0.09	

A stepwise regression of all sediment variables on %MeHg explained about 40% of the residual variability in MeHg in sediments. The two variables that best predicted %MeHg were reduced sulfur (CRS) (negative relationship) and the organic matter content of sediments (positive relationship). When the model was expanded to include land use, Hg deposition, watershed and hydrologic parameters, and water chemistry, no other variables entered the model.

A model examining %MeHg in sediments vs. water chemistry, land use, hydrology and morphology was constructed, using the average data for all 14 reservoirs. Only water depth entered the model.

Model for %MeHg in sediments (all non-sediment parameters)		
model logsedperMeHg = age depth logsurfarea logcap logwatarea logsurftowat logflow restime HgdepDNR logLUdev logLUag logLUfor logLUwet ph logTSS logDOC logSO4 logbottDO /selection = stepwise;		
DF	13	
Model r^2	0.27	
Model p	0.055	
Variables included	***Partial r^2***	***p***
depth	0.27	0.055

Water. Across all variables, the best predictor of MeHg in the water column (other than total Hg) was pH (negative).

Model for MeHg in water (all variables)		
model logwatperMeHg = ph logTSS logDOC logCl logNO3 logSO4 logbottDO/selection = stepwise;		
DF	12	
Model r^2	0.903	
Model p	0.0004	
Variables included	***Partial r^2***	***p***
pH	0.46	<0.0001
logSO4	0.26	0.0005
logLUag	0.11	0.005

Individual correlations among variables showed that water column % MeHg was strongly positively related to both Hg and MeHg concentrations, and weakly negatively correlated with pH and chloride.

For the stepwise linear regression analysis, the average water column data set for all eastern and western reservoirs was used (Tables 16, 18 and 19), except Duckett reservoir, which was excluded from the analysis (n=13). A stepwise linear regression model of %MeHg on all water, sediment, land use and hydrologic variables showed pH, and sulfate as the most significant variables, followed by percent agricultural land use (all positive).

Model for %MeHg in water (non-Hg all variables)		
model logwatperMeHg = age depth logsurfarea logsurftowat logflow restime logLUdev logLUag logLUfor logLUwet ph logTSS logDOC logSO4 logbottDO loi		
DF	12	
Model r^2	0.98	
Model p	<0.0001	
Variables included	***Partial r^2***	***p***
pH (neg)	0.46	0.011
logSO4	0.26	0.013
logLUag	0.11	0.036
logLUdev	0.07	0.043

Factors related to bioaccumulation in fish. The ratio of MeHg in fish to MeHg (filterable) in water is the bioaccumulation factor (BAF), and is commonly at least 1 million for mature predatory fish. The BAF for size-normalized largemouth bass varied by almost an order of magnitude across the 14 reservoirs studied (Figure 28), and generally increased from east to west. The calculated BAFs were based on the 2000-2005 average water column MeHg concentrations given in Table 18 and the largemouth bass collected in 2000-2001. Parameters that were individually correlated with BAF ($P<0.1$) were bottom water DO (positive correlation); lake surface:watershed area ratio (positive correlation), percent developed land (negative), pH (positive) and TSS (negative).

A stepwise linear regression model of BAF on non-Hg water chemistry parameters included bottom water dissolved oxygen (positive relationship) and pH (also positive) as significant variables.

Model for BAF (water chemistry only)		
model logLMBBAF = pH logTSS logDOC logSO4 logbottDO		
DF	12	
Model r^2		
Model p	0.012	
Variables included	***Partial r^2***	***p***
logbottDO	0.42	0.013
pH	0.11	0.15

Expanded models that included all non-Hg variables (land use variables, watershed physical and hydrologic parameters, and sediment variables), brought in lake surface:watershed area ratios (positive) and percent wetland area (positive).

MeHg in fish can derive from MeHg in water, sediment or both. The relationship between BAF and surface area to volume ratio suggests that sediments are contributing MeHg directly to fish in these reservoirs. Water bodies with higher surface to volume ratios also have higher sediment surface area to volume ratios.

Overall model for Hg in largemouth bass. Models for Hg in fish were based on size-normalized concentrations in a 370 mm fish (data from Sveinsdottir and Mason, 2005). The best overall predictor of Hg in largemouth bass was filterable MeHg in the water column (Figure 29). Stepwise linear regression analysis for the 14 lake data set, including all variables, gave dissolved MeHg concentrations, the lake's surface to water ratio, and the percentage of wetlands in the watershed as significantly predictive variables.

Model for Hg in largemouth bass (all parameters)		
model LMB = age depth logsurfarea logcap logwatarea logsurftowat logflow restime HgdepDNR logLUdev logLUag logLUfor logLUwet logHgD logHgUNF logMeHgD logMeHgUNF ph logTSS logDOC logSO4 logbottDO		
DF	12	
Model r^2	0.78	
Model p	0.0025	
Variables included	***Partial r^2***	***p***
logMeHgD	0.55	0.0003
logsurftowat	0.14	0.016
logLUwet	0.1	0.078

A stepwise regression model that excluded all sediment variables, and all Hg concentration parameters (n=14) gave pH (negative) as the most significant variable.

Model for Hg in largemouth bass (w/o sediment and Hg data)		
model LMB = age depth logsurfarea logcap logwatarea logsurftowat logflow restime HgdepDNR logLUdev logLUag logLUfor logLUwet ph logTSS logDOC logSO4 logbottDO /selection = stepwise;		
DF	13	
Model r^2	0.60	
Model p	0.01	
Variables included	***Partial r^2***	***p***
pH	0.48	0.009
logbottDO	0.12	0.11

Table 16. Final compiled data set for statistical modeling, part A. Length-normalized (370 mm) largemouth bass Hg data (from Sveinsdottir and Mason 2005) in ng/g wet weight. The BAF was calculated from the LMB data in this table, and the average water column MeHg values given in Table 18.

Reservoir	LMB length weighted avg Hg (ng/g)	LMB BAF
Clopper	218	1.03E+06
DeepCreek	308	1.07E+06
Duckett	222	2.07E+06
Lariat	643	1.06E+06
Liberty	277	1.98E+06
LochRaven	304	8.96E+05
PineyFrostburg	607	4.12E+06
PineyRunLake	156	7.82E+05
PrettyBoy	335	2.67E+06
RockyGap	108	1.80E+06
Savage	521	7.15E+06
StMarysLake	776	5.08E+05
Triadelphia	174	1.37E+06
Tuckahoe	323	2.62E+06

Table 17. Data used to calculate surface water Hg and MeHg concentrations for statistical modeling. Data from 2003-2005 from this study. Data from 2001-2002 from Svensdottir and Mason 2005.

SURFACE WATER DATA	2003-2005				2000-2001				AVG USED IN MODELLING			
Reservoir	**HgT (ng/L) Whole**	**HgT (ng/L) Filtered**	**MeHg (pg/L) Whole**	**MeHg (pg/L) Filtered**	**HgT (ng/L) Whole**	**HgT (ng/L) Filtered**	**MeHg (pg/L) Whole**	**MeHg (pg/L) Filtered**	**HgT (ng/L) Whole**	**HgT (ng/L) Filtered**	**MeHg (pg/L) Whole**	**MeHg (pg/L) Filtered**
Clopper	2.60	0.66	0.22	0.12	16.99	18.05	0.226	0.094	9.79	9.35	0.22	0.11
Deep Creek	0.33	0.21	0.034		2.42	0.62	0.393	0.285	1.37	0.41	0.21	0.28
Duckett Res.	18.04	35.02	0.18	0.13	15.39	19.53	0.115	0.050	16.72	27.28	0.15	0.09
Lake Lariat	2.74	1.89	1.59	1.10	2.42	2.10	0.123	0.016	2.58	1.99	0.86	0.56
Liberty	0.51	0.55	0.14	0.16	1.99	1.89	0.064	0.075	1.25	1.22	0.10	0.12
Loch Raven	0.60	0.45	0.37	0.15	5.09	5.63	0.195	0.160	2.84	3.04	0.28	0.15
Piney Res	0.84	0.54	0.038		2.78	1.41	0.337	0.262	1.81	0.97	0.19	0.26
Piney Run Lake	0.70	0.43	0.00	0.04	1.69	0.97	0.337	0.255	1.19	0.70	0.17	0.15
Pretty Boy	1.15	0.47	0.21	0.09	3.95	4.03	0.047	0.058	2.55	2.25	0.13	0.08
Rocky Gap	0.63	0.45	0.019		0.40		0.129	0.036	0.51	0.45	0.07	0.04
Savage	0.55	0.34	0.069		1.28	0.61	0.105	0.073	0.92	0.47	0.09	0.07
St Mary's Lake	6.17	0.84	0.34	0.21	2.17	0.46	0.190	0.131	4.17	0.65	0.27	0.17
Triadelphia Res.	6.71	0.38	0.04	0.09	2.67	1.96	0.076	0.076	4.69	1.17	0.06	0.08
Tuckahoe	1.15	0.56	0.10	0.04	4.08	2.40	0.126	0.137	2.61	1.48	0.11	0.09

Table 18. Whole water column Hg and MeHg averages used for statistical modeling. Values were calculated as the (average of all surface water data) + (average of all bottom water data)/2. Data used to derive average surface water data are shown in Table 17. Bottom water data for 2003-2005 are in Table 19. Limited bottom water concentration data were also available for 2001-2002.

Reservoir	HgT (ng/L) Whole	HgT (ng/L) Filtered	MeHg (ng/L) Whole	MeHg (ng/L) Filtered
Clopper	5.63	4.93	0.21	0.12
DeepCreek	1.13	0.45	0.29	0.27
Duckett	31.51	31.50	0.11	0.06
Lariat	2.28	1.78	0.61	0.40
Liberty	1.30	0.85	0.14	0.11
LochRaven	1.83	1.95	0.34	0.12
PineyFrostburg	1.50	0.84	0.15	0.26
PineyRunLake	1.52	0.74	0.20	0.17
PrettyBoy	2.31	1.36	0.13	0.07
RockyGap	0.66	0.44	0.06	0.05
Savage	0.79	0.43	0.07	0.07
StMarysLake	3.72	1.25	1.53	1.04
Triadelphia	3.11	0.89	0.13	0.07
Tuckahoe	1.69	1.03	0.12	0.09

Table 19. Water chemistry data used in statistical modeling. Values are averages of data collected in 2003-2005 (this study) and 2001-2002 (Svensdottir and Mason 2005).

Reservoir	pH	TSS (mg/L)	DOC	Chloride (mg/l)	Nitrate (mg/l)	Sulfate (mg/l)	Bottom water DO
Clopper	7.40	4.17	7.12	130.19	0.37	4.50	0.48
Deep Creek	6.84	2.32	2.66	7.67	0.05	12.91	0.44
Duckett Reservoir	6.84	9.79	2.77	61.37		6.26	0.77
Lake Lariat	6.49	7.75	4.14	20.70	0.53	9.30	0.17
Liberty	7.06	2.32	10.57	14.91	1.96	5.28	0.48
Loch Raven	7.58	27.90	6.79	49.37	1.77	3.87	0.15
Piney (Frostburg Reservoir)	6.98	1.74	4.31	24.18	0.81	7.49	2.25
Piney Run Lake	7.20	5.65	2.89	27.96	0.58	5.99	0.00
Pretty Boy	7.30	7.61	10.44	23.78	1.89	2.61	0.17
Rocky Gap (Lake Habeeb)	7.51	1.99	3.77	3.29	0.04	10.27	3.99
Savage	7.22	3.92	2.03	10.70	0.58	11.49	3.82
St Mary's Lake	4.84	14.56	9.06	9.00	6.51	3.95	0.25
Triadelphia Reservoir	6.71	21.74	3.38			5.55	0.04
Tuckahoe	7.47	2.50	8.47	15.76	2.82	5.57	5.44

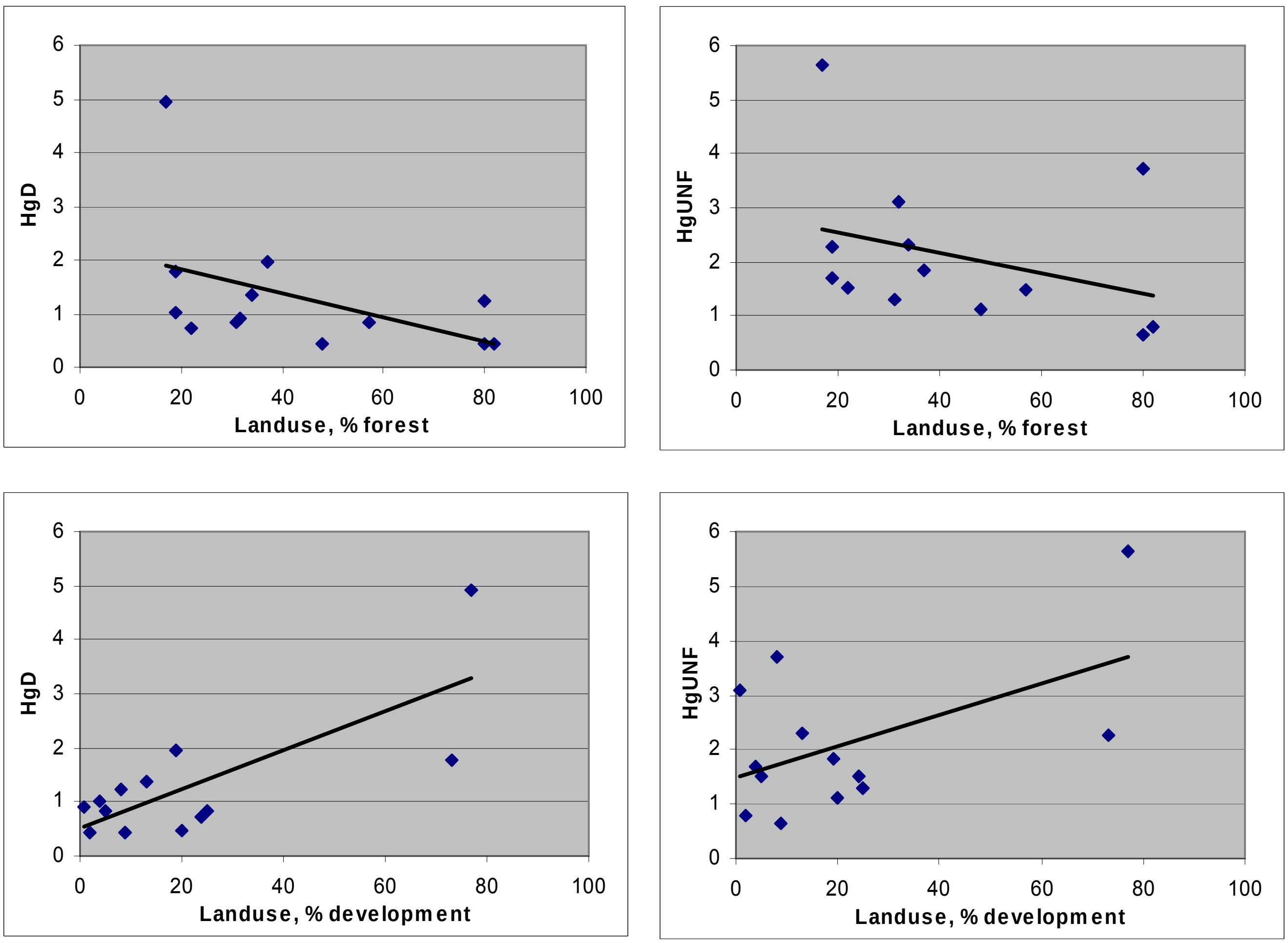
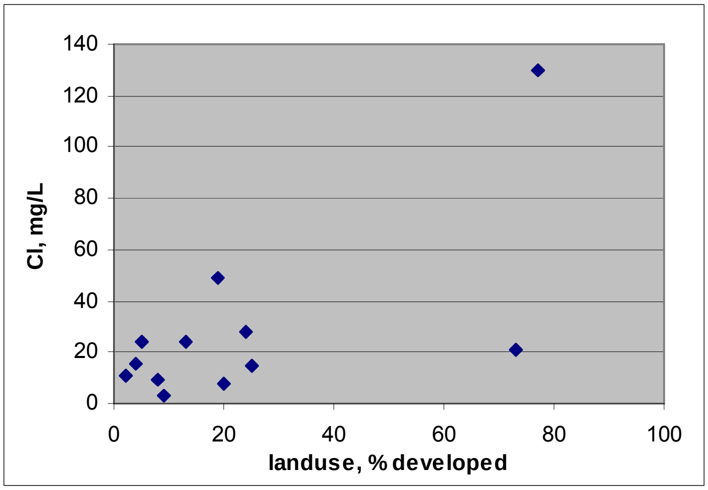
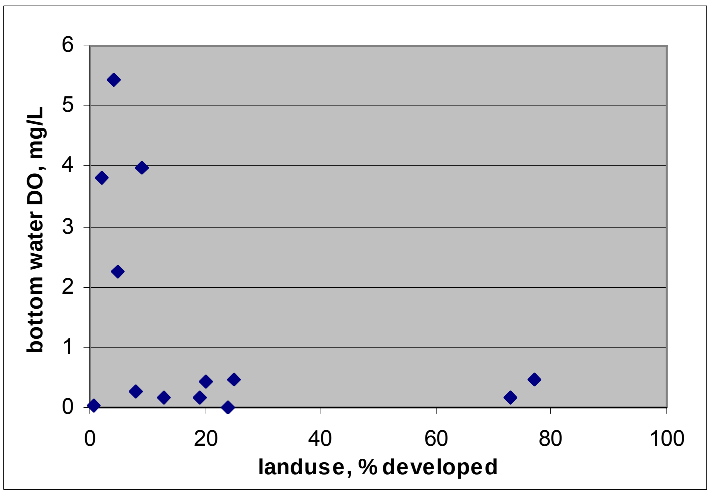


Figure 26. The relationship between filterable (left) and total (right) Hg surface waters and land use for 13 reservoirs.





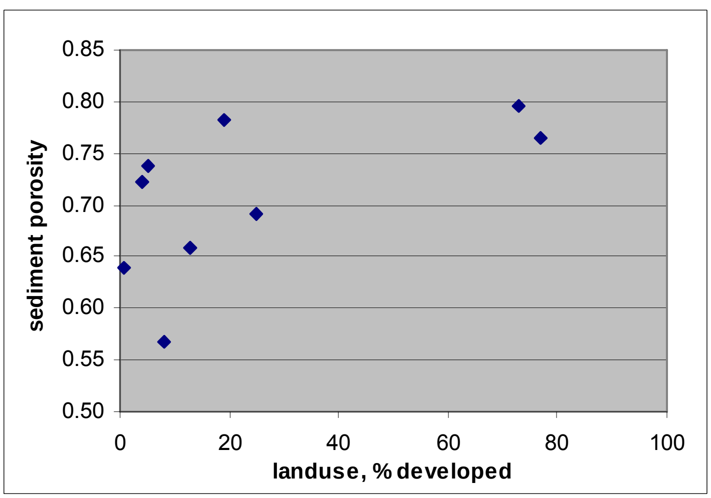


Figure 27. Signficant relationships between land use and sediment and water chemistry.

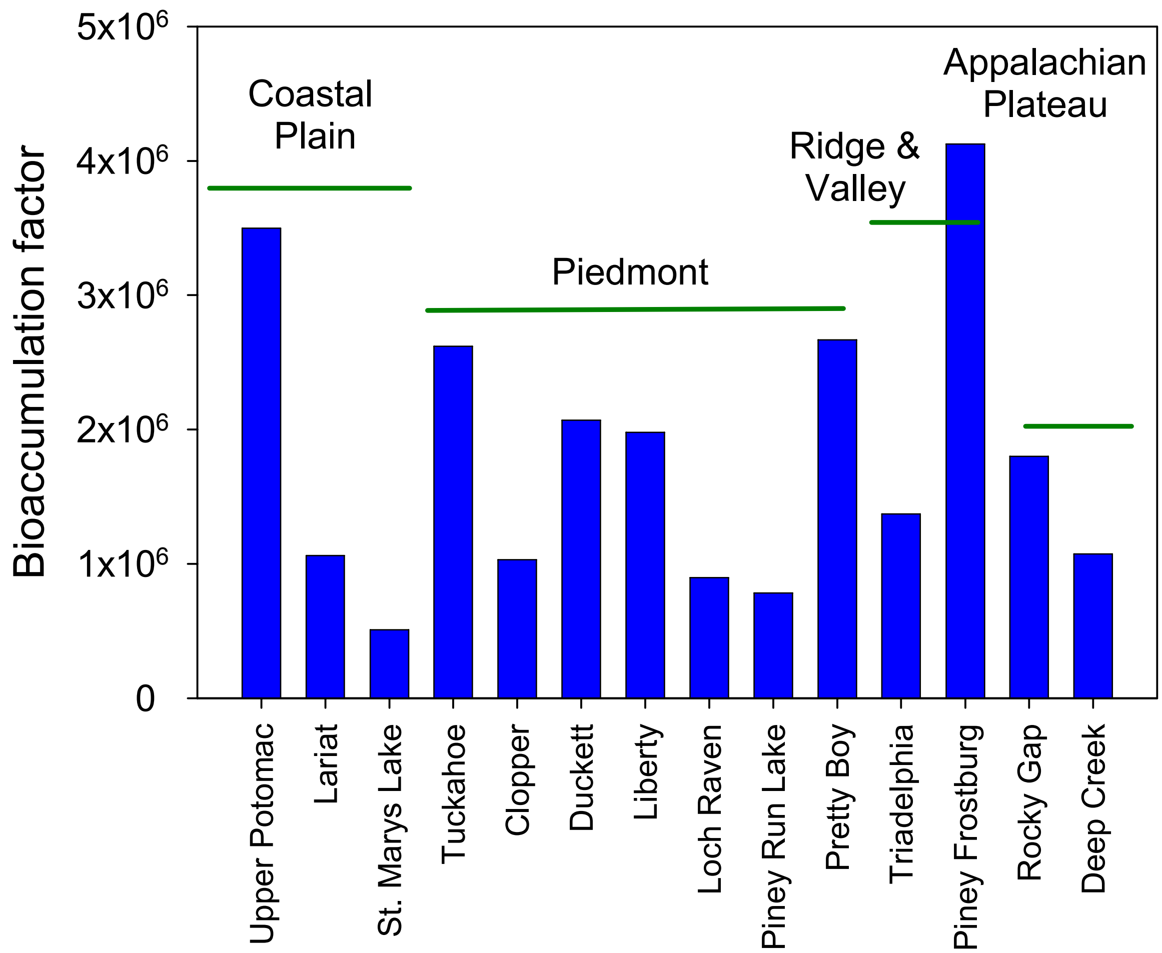


Figure 28. Bioaccumulation factors for size –normalized largemouth bass, where BAF = MeHg (mg/kg) in largemouth bass / filterable MeHg in water (mg/L).

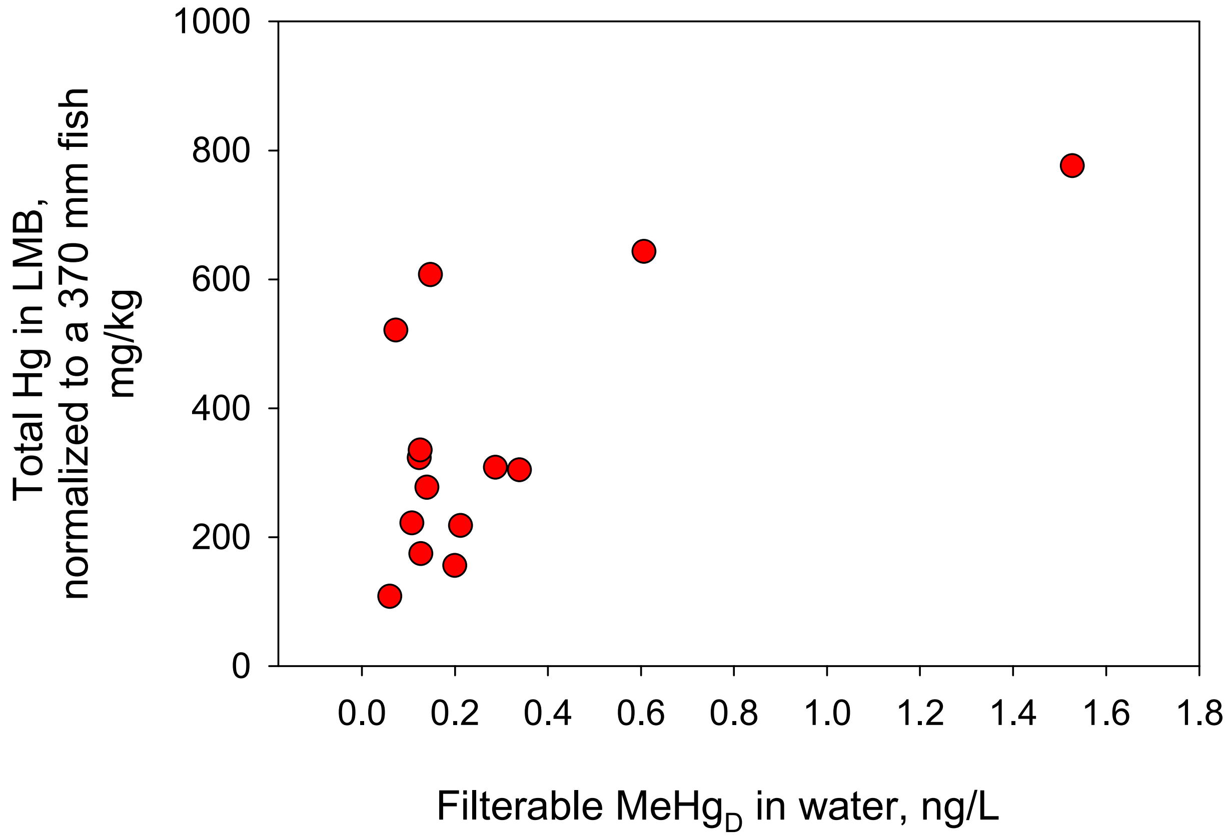


Figure 29. The relationship between water column filterable MeHg concentrations and Hg concentrations largemouth bass, normalized to a 370 mm fish. .

Discussion and Conclusions

Size- and species- normalized Hg levels in fish vary by almost a factor of 10 across the 14 Maryland reservoirs examined in this study. Our objective was to determine the factors that most strongly correlate with Hg levels in fish, in order to aid managers in choosing effective Hg management practices. A large body of cause and effect research on the controls on Hg in fish led us to choose the variables examined.

Rather than a statistical analysis that examined Hg concentrations in fish against all potential controlling variables, we chose to examine each of the major steps in the Hg cycle separately. Statistical analyses of similar data sets in other regions have not been approached this way. Because of the complexity of the Hg cycle, the large number of variables that affect Hg levels in fish, and the relatively small number of reservoirs examined, this approach provided more power to assess potential controls on Hg bioaccumulation.

A summary of results is shown below. Stepwise regressions models for Hg in largemouth bass revealed a very strong correlation with MeHg levels in water, and pH, but little more. Models for each component of the Hg cycle revealed the sequential controls on bioaccumulation. Land use, water and sediment chemistry and Hg deposition explained most of the variability in Hg in sediments and water. In turn, Hg concentrations in sediment and water, along with pH, sulfate, DO, and organic matter, were the best predictors of MeHg in sediment and water. The bioaccumulation of MeHg from water to fish was related to DO, pH and the reservoir surface to water ratio.

Land use, particularly the percent developed land, accounted for about 35% of the variability of Hg in water. One likely explanation is enhanced transport of atmospherically deposited Hg across impervious surfaces; another is direct Hg contributions from developed landscapes. Water column Hg concentrations dropped dramatically with increasing percent forested land in the watershed. The potential role of forested buffers in minimizing Hg transport to receiving waters should be investigated as a control mechanism for Hg in fish.

Water chemistry, specifically chloride and DOC concentrations, accounted for significant additional variability in Hg in water. Land use also impacted chloride concentrations in surface waters. Buffers against runoff could potentially limit both Hg and chloride inputs to reservoirs. The relationship between chloride and Hg may be incidental – both driven by land use – or chloride may be acting as a ligand to hold Hg in solution.

Importantly, Hg deposition rates explained a significant portion of the variability in water column Hg concentrations, after land use and water chemistry were accounted for. The variability in sediment Hg was driven by the grain size and organic matter content of sediments, but Hg deposition rates also contributed.

The major control on MeHg in both sediment and water appears to be the inorganic Hg concentration. Importantly, most of the Hg in US sediments derives from anthropogenic sources (Kamman and Engstrom 2002; Engstrom et al. 2007), probably nearly all of it from direct and indirect atmospheric deposition (Lindberg et al. 2007). Sulfate and pH accounted for significant additional variability in water column MeHg. Sulfate is known to stimulate MeHg production through the action of sulfate-reducing bacteria. Acidity is also commonly identified as a correlate of MeHg in aquatic ecosystems, affecting methylation, partitioning, and bioaccumulation. These relationships support the commonly held contention that reduction in acid deposition to freshwater ecosystems – particularly sulfates – will reduce the net production of MeHg from inorganic Hg. In a separate analysis that included surface and bottom water data rather than lake averages, low DO in bottom waters was strongly correlated with MeHg.

Coastal Plain reservoirs seem particularly sensitive to Hg. This appears to be a result of high rates of net MeHg production, driven by low pH, and low DO, and relatively reduced sediments. Flux of MeHg from watersheds could also potentially contribute, but this was not assessed here. Bioaccumulation factors are relatively low for these systems.

The bioaccumulation of MeHg from water into fish was also correlated with dissolved oxygen. Reservoirs with low or zero DO bottom water had generally higher BAFs. Turnover of high MeHg bottom waters into surface waters in the fall may increase MeHg levels in water well above those measured in the summer. A positive relationship between BAF and surface to water area ratios suggest direct MeHg uptake from sediments.

Stepwise regression of all variables on largemouth bass Hg concentrations gave MeHg in water as the most important driver, followed by pH.

Table 20. Summary of results from statistical analyses.

Component Process in Hg cycle	**Dependent variables assessed**	**Significantly correlated variables**
Hg deposition, transport and accumulation in lakes	Hg in water	• Water chemistry – Cl (+), DOC (+), SO4 (+) • Land use: % developed (+), % forested (-) • Hg deposition rates (+) • Reservoir surface area (+)
	Hg in sediment	• Sediment grain size/organic matter content • Hg in water (+) • Land use: % developed land (+) • Hg deposition rates (+)
Net MeHg production	% MeHg in water	• pH (-) • SO4 (+) • Bottom water DO (-) • Land use: % ag or developed land (+)
	% MeHg in sediment	• Reduced sulfide (-) • Organic matter content (+)
MeHg bioaccumulation	Bioaccumulation factors for size-normalized largemouth bass	• Dissolved oxygen in bottom water (-) • pH (+) • Lake surface:water ratio
Overall	Size-normalized Hg in largemouth bass	• MeHg in water (+) • Lake surface:water ratio • pH (-)

Recommendations

Investigate the potential role of forested buffers and porous surfaces in minimizing Hg transport to receiving waters. There appear to be multiple negative aspects of developed landscapes on Hg.

Reduce Hg emissions in Maryland. Recent models by MD DNR suggest that a substantial fraction of Hg deposition in Maryland derives from Maryland sources. Mercury deposition, along with developed landscapes, appear to be the major drivers of Hg levels in Maryland reservoirs.

Improve understanding of “dry deposition,” in order to improve understanding of total Hg deposition rates, mechanisms, sources and remediation. Data from other regions suggest that dry deposition may equal or exceed wet deposition for Hg. Wet deposition rates are now being monitored in Maryland, but techniques for adequately measuring dry deposition rates are still being developed.

Reduce emissions of SO_X in Maryland. Minimize acid mine drainage to western reservoirs. Sulfate and pH are important drivers of MeHg production in Maryland.

Monitor the impacts of Hg and SOx emissions regulations on Hg deposition and Hg bioaccumulation. Both the timing and magnitude of change should be considered in order to assess the effectiveness of new regulations. Monitoring should begin as soon as possible so that some baseline can be established prior to implementation of new regulations.

Repeat water column sampling in T.H Duckett reservoir. Anomalously high inorganic Hg levels were found in samplings in two different years in Duckett Reservoir.

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Appendix I. An Examination of the Factors that Control Methylmercury Production and Bioaccumulation in Maryland Reservoirs

Full correlation matrix

RAW DATA

Obs	Reservoir	LMB	LMBBAF	Age	Depth	SurfArea	Cap
1	Clopper	218	1030000	31	12	0.36	2.00E+06
2	DeepCreek	308	1070000	81	25	18	1.10E+08
3	Duckett	222	2070000	53	22.6	3.13	2.10E+07
4	Lariat	643	1060000	41	9.1	0.39	1.90E+06
5	Liberty	277	1980000	53	44	12.57	1.60E+08
6	LochRaven	304	896000	83	23.2	9.71	9.00E+07
7	PineyRunLake	607	4120000	72	9.8	0.49	1.70E+06
8	PineyFrostburg	156	782000	16	54.5	1.21	9.56E+06
9	PrettyBoy	335	2670000	70	30	6.07	7.40E+07
10	RockyGap	108	1800000	37	25	0.85	6.60E+06
11	Savage	521	7150000	54	46.1	1.46	2.50E+07
12	StMarysLake	776	508000	31	6.4	1.01	3.90E+06
13	Triadelphia	174	1370000	63	15.8	3.24	2.30E+07
14	Tuckahoe	323	2620000	31	2.7	0.35	3.20E+04

Obs	WatArea	SurftoWat	Flow	ResTime	HgDep	HgDep DNR	Ludev
1	7	19	0.17	0.38	2.81	24	77
2	163	9	8.66	0.4	0.97	29	20
3	343	110	2.38	0.28	0.24	27	2
4	7	18	0.09	0.71	0.37	18	73
5	44	34	5.46	0.93	0.17	28	25
6	789	81	8.6	0.33	0.65	37	19
7	31	63	0.65	0.08	0.16	27	5
8	8	23	0.36	0.84	0.16	26	24
9	6	34	2.9	0.81	0.13	33	13
10	3	27	0.34	0.62	0.1	22	9
11	70	185	4.17	0.19	0.3	29	2
12		22	0.26	0.48	0.33	18	8
13	5	63	2.35	0.31	0.23	28	1
14	3	640	2.78	0.000365	0.09	20	4

Obs	Luag	Lufor	Luwet	HgUNF	HgD	MeHg UNF	MeHgD
1	1	17	1	5.63	4.93	0.21	0.12
2	0	48	5	1.13	0.45	0.29	0.27
3	56	37	3	31.51	31.5	0.11	0.06
4	3	19	4	2.28	1.78	0.61	0.4
5	41	31	1	1.3	0.85	0.14	0.11
6	4	37	1	1.83	1.95	0.34	0.12
7	37	57	0	1.5	0.84	0.15	0.26
8	50	22	2	1.52	0.74	0.2	0.17
9	48	34	0	2.31	1.36	0.13	0.07
10	7	80	0	0.66	0.44	0.06	0.05
11	15	82	1	0.79	0.43	0.07	0.07
12	8	79	5	3.72	1.25	1.53	1.04
13	63	32	3	3.11	0.89	0.13	0.07
14	61	19	15	1.69	1.03	0.12	0.09

Obs	watper MeHg	pH	TSS	DOC	Cl	NO3	SO4
1	0.46	7.4	4.17	7.12	130.19	0.37	4.5
2	60.39	6.84	2.32	2.66	7.67	0.05	12.91
3	0	6.84	9.79	2.77	61.37	.	6.26
4	0.7	6.49	7.75	4.14	20.7	0.53	9.3
5	13.13	7.06	2.32	10.57	14.91	1.96	5.28
6	6.3	7.58	27.9	6.79	49.37	1.77	3.87
7	31.1	6.98	1.74	4.31	24.18	0.81	7.49
8	3	7.2	5.65	2.89	27.96	0.58	5.99
9	5.5	7.3	7.61	10.44	23.78	1.89	2.61
10	1.1	7.51	1.99	3.77	3.29	0.04	10.27
11	17.11	7.22	3.92	2.03	10.7	0.58	11.49
12	83.77	4.84	14.56	9.06	9	6.51	3.95
13	7.79	6.71	21.74	3.38	.	.	5.55
14	9	7.47	2.5	8.47	15.76	2.82	5.57

Obs	BottDO	pwHg	pw MeHg	pwper MeHg	pwHS	pwFe	pwMn
1	0.48	1.64	0.24	11.57	4.09	16.68	0.04
2	0.44	.	.	.	.	.	.
3	0.77	1.98	0.17	9.18	0.03	28.87	10.1
4	0.17	1.36	0.29	24.29	0	850.98	1.46
5	0.48	1.91	0.93	48.22	.	36.98	0.11
6	0.15	0.89	0.1	10.79	0	14.41	0.07
7	0.5	0.96	0.02	3.82	0	19.93	4.1
8	0	.	.	.	.	.	.
9	0.17	2.2	0.52	24.56	0.14	27.82	0.1
10	3.99	.	.	.	.	.	.
11	3.8	.	.	.	.	.	.
12	0.5	3.73	0.34	9.27	0.01	1320.93	1.77
13	0.04	2.33	0.48	19.38	0	15.74	4.16
14	5.44	0.81	0.11	12.58	0	12.54	0.03

Obs	pwDOC	Bulkden	dw	Por	LOI	AVS	CRS
1	9.96	1.16	0.276	0.764	8.65	27.72	37.92
2	.	.	.	.	9.31	.	.
3	0.3	1.3	0.553	0.604	6.42	2.95	13.07
4	16.44	1.08	0.222	0.796	12.46	192.28	194.76
5	17.64	1.25	0.393	0.692	10.07	2.53	9.31
6	10.63	1.11	0.256	0.782	14.66	70.9	107.07
7	17.58	1.12	0.306	0.739	8.22	11.7	40.22
8	.	.	.	.	9.71	.	.
9	17.5	1.26	0.436	0.658	9.07	0.47	20.7
10	.	.	.	.	8.02	.	.
11	.	.	.	.	8.35	.	.
12	6.13	1.3	0.582	0.567	5.16	13.68	78.58
13	16.87	1.26	0.475	0.638	6.88	3.39	12.24
14	11.55	1.19	0.349	0.723	17.91	2.37	22.12

Obs	FeII	FeIII	sedHg	sedMeHg	sedper	KdHg	KdMeHg
1	5.01	0.05	99.83	0.87	0.74	71800	4700
2	.	.	64.07	0.44	0.76	.	.
3	16.7	0.42	58.42	0.43	0.7	27600	2450
4	0.89	0.17	95.12	1.09	1.17	78300	5590
5	1	0.05	87.51	1.07	1.35	53700	1390
6	45.5	0.09	209.38	1.62	0.79	304000	16200
7	4.75	0.43	62.59	0.38	0.58	76100	12000
8	.	.	65.74	0.57	0.95	.	.
9	10	0.26	73.41	0.71	1.34	49300	1600
10	.	.	33.82	0.15	0.68	.	.
11	.	.	45.61	1.2	2.61	.	.
12	9.69	1.79	53.09	0.42	0.83	13600	1230
13	9	0.19	46.28	0.54	1.1	20300	1320
14	7.9	0.14	95.4	0.75	0.86	151000	16800

Variable statistics and units

Variable	N	Mean	Std Dev	Minimum	Maximum	Units	Description
watperMeHg	14	21.06	6.34	0.2	83.77	%	avg % MeHg/Hg in lake water column
sedperMeHg	14	1.03	0.14	0.58	2.61	%	avg % MeHg/Hg in surface sediments
LMB	14	355.14	54.14	108	776	ug/kgww	length-weighted Hg concentration in largemouth bass, ug/kg
LMBBAF	14	2.08E+06	4.68E+05	5.08E+05	7.15E+06	ratio	largemouth bass bioaccumulation factor (g Hg/Kg fish)/(g Hg/L water)
Age	14	51.14	5.55	16	83	y	reservoir age
Depth	14	23.30	4.23	2.7	54.5	m	normal maximum depth
SurfArea	14	4.20	1.47	0.35	18	m2	lake surface area
Cap	14	3.78E+07	1.35E+07	3.20E+04	1.60E+08	m3	Normal capacity
WatArea	14	195.79	58.36	7	789	km2	Watershed area
SurftoWat	14	94.86	43.81	9	640	ratio	lake surface/watershed area
Flow	14	2.80	0.79	0.09	8.66	m3/s	avg annual flow
ResTime	14	0.45	0.08	0.000365	0.93	y	Hydraulic residence time
HgDepDNR	14	26.14	1.45	18	37	ug/m2 y	total annual wet deposition from MD DNR deposition model
Ludev	14	20.14	6.59	1	77	%	Landuse, % developed
Luag	14	32.29	6.02	1	63	%	Landuse, % agricultural
Lufor	14	42.43	6.24	17	82	%	Landuse, % forest
Luwet	14	2.93	1.04	0	15	%	Landuse, % wetland
HgD	14	3.46	2.18	0.43	31.5	ng/L	avg filterable Hg concentration in the water column
HgUNF	14	4.21	2.13	0.66	31.51	ng/L	avg total Hg concentration in the water column
MeHgD	14	0.21	0.07	0.05	1.04	ng/L	avg filterable MeHg concentration in the water column
MeHgUNF	14	0.29	0.10	0.06	1.53	ng/L	avg total MeHg concentration in the water column
pH	14	6.96	0.18	4.84	7.58	-log[H+]	avg water column pH
TSS	14	8.14	2.15	1.74	27.9	mg/L	avg total suspended solids in the water column
DOC	14	5.60	0.81	2.03	10.57	mg/L	dissolved organic carbon
Cl	13	30.68	9.48	3.29	130.19	mg/L	filterable chloride
NO3	1	1.49	0.52	0.04	6.51	mg/L	filterable nitrate
SO4	14	6.79	0.83	2.61	12.91	mg/L	filterable sulfate
BottDO	14	1.32	0.48	0	5.44	mg/L	Hydrolab
pwHg	10	1.78	0.28	0.81	3.73	ng/L	avg filterable Hg concentration in0-4 cm depth sediment pore waters
pwMeHg	10	0.32	0.09	0.02	0.93	ng/L	avg filterable MeHg concentration in0-4 cm depth sediment pore waters
pwFe	10	234.49	146.19	12.54	1320.93	mg/L	avg filterable total Fe concentration in0-4 cm depth sediment pore waters
pwMn	10	2.19	1.02	0.03	10.1	mg/L	avg filterable total Mn concentration in0-4 cm depth sediment pore waters
pwDOC	10	16.46	1.54	9.96	26.13	mg/L	avg DOC concentration in0-4 cm depth sediment pore waters
Bulkden	10	1.20	0.03	1.08	1.3	gww/cm3	sediment bulk density
Por	10	0.70	0.02	0.567	0.796	ml/cm3	sediment porosity
dw	10	0.38	0.04	0.222	0.582	gdw/cm3	sediment dry weight
LOI	14	9.64	0.90	5.16	17.91	%	sediment loss on ignition
FeII	10	18.12	3.64	7.92	45.25	mg/gdw	Extractable Fe(II), 0-4 cm sediments
FeIII	10	0.36	0.16	0.05	1.79	mg/gdw	Extractable Fe(III), 0-4 cm sediments
sedHg	14	77.88	11.49	33.82	209.38	ng/gdw	Total Hg in 0-4 cm bulk sediments
sedMeHg	14	0.73	0.11	0.15	1.62	ng/gdw	MeHg in 0-4 cm bulk sediments
AVS	10	32.80	18.96	0.47	192.28	umoles/gdw	acid volatile sulfides
CRS	10	53.60	18.63	9.31	194.76	umoles/gdw	chromium-reducible sulfides
KdHg	10	8.46E+04	2.74E+04	1.36E+04	3.04E+05	(ng/kg)/(ng/L)	solid:aqueous partition coeficient for Hg in sediments
KdMeHg	10	6.33E+03	1.99E+03	1.23E+03	1.68E+04	(ng/kg)/(ng/L)	solid:aqueous partition coeficient for MeHg in sediments

Transformed data used in correlation matrix:

45 Variables	LMB	logLMBBAF	Age	Depth	logsurfarea	logcap
	logwatarea	logsurfto	wat logflow	ResTime	logHgdep	logLUdev
	logLUag	logLUfor	logLUwet	logHgD	logHgUNF	logMeHgD
	logMeHgUNF	pH	logTSS	logDOC	logCl	logNO3
	logSO4	logbottdo	logpwHg	logpwMeHg	log2pwFe	logpwMn
	pwDOC	Bulkden	Por	dw	LOI	logFeII
	logFeIII	logsedHg	logsedMeHg	logAVS	logCRS	logkdHg
	logkdMeHg	logwatperMeHg	sedperMeHg			

Simple statistics

Variable	N	Mean	Std Dev	Minimum	Maximum
logwatperMeHg	14	2.37	1.48	-1.61	4.43
sedperMeHg	14	1.03	0.51	0.58	2.61
LMB	14	355.14	202.56	108.00	776.00
logLMBBAF	14	14.30	0.71	13.14	15.78
Age	14	51.14	20.77	16.00	83.00
Depth	14	23.30	15.81	2.70	54.50
logsurfarea	14	0.61	1.37	-1.05	2.89
logcap	14	16.09	2.27	10.37	18.89
logwatarea	14	4.44	1.57	1.95	6.67
logsurftowat	14	3.83	1.11	2.20	6.46
logflow	14	0.23	1.53	-2.41	2.16
ResTime	14	0.45	0.29	0.00	0.93
logHgdepDNR	14	3.24	0.21	2.89	3.61
logLUdev	14	2.30	1.32	0.00	4.34
logLUag	14	3.00	1.28	0.00	4.14
logLUfor	14	3.61	0.55	2.83	4.41
logLUwet	11	0.93	0.89	0.00	2.71
logHgD	14	0.26	1.13	-0.84	3.45
logHgUNF	14	0.78	0.96	-0.42	3.45
logMeHgD	14	-2.00	0.85	-3.00	0.04
logMeHgUNF	14	-1.66	0.86	-2.81	0.43
pH	14	6.96	0.69	4.84	7.58
logTSS	14	1.70	0.91	0.55	3.33
logDOC	14	1.58	0.56	0.71	2.36
logCl	13	2.99	0.96	1.19	4.87
logNO3	1	-0.36	1.52	-3.22	1.87
logSO4	14	1.82	0.46	0.96	2.56
logbottdo	13	-0.57	1.50	-3.22	1.69
logpwHg	10	0.47	0.49	-0.21	1.32
logpwMeHg	10	-1.54	1.09	-3.91	-0.07
log2pwFe	10	1.26	0.37	0.93	1.97
logpwMn	10	-0.78	2.20	-3.51	2.31
pwDOC	10	16.46	4.86	9.96	26.13
Bulkden	10	1.20	0.08	1.08	1.30

Correlation matrix
P values highlighted
Italic: *P<0.05*
Bold: **P<0.5**

		logwatper MeHg	sedper MeHg	LMB	log LMBBAF	Age	Depth	logsurfarea	logcap	logwatarea	logsurf towat	logflow	ResTime	log HgdepDNR
logwatperMeHg	r	1												
	p													
	n	14												
sedperMeHg	r	0.11225	1											
	p	0.7024												
	n	14	14											
LMB	r	0.50678	0.20479	1										
	p	**0.0644**	0.4825											
	n	14	14	14										
logLMBBAF	r	-0.19067	0.53987	0.03155	1									
	p	0.5138	*0.0463*	0.9147										
	n	14	14	14	14									
Age	r	-0.02467	0.04112	0.04163	0.27243	1								
	p	0.9333	0.889	0.8876	0.346									
	n	14	14	14	14	14								
Depth	r	0.01092	0.5213	-0.37502	0.18937	-0.02056	1							
	p	0.9705	0.0559	0.1864	0.5167	0.9444								
	n	14	14	14	14	14	14							
logsurfarea	r	-0.04862	0.11184	-0.29333	-0.07736	0.66538	0.46064	1						
	p	0.8689	0.7035	0.3088	0.7926	*0.0094*	**0.0974**							
	n	14	14	14	14	14	14	14						
logcap	r	-0.05411	0.25965	-0.23164	-0.05946	0.57601	0.61462	0.8773	1					
	p	0.8542	0.37	0.4256	0.84	*0.0311*	*0.0193*	*<.0001*						
	n	14	14	14	14	14	14	14	14					
logwatarea	r	-0.2925	0.28455	-0.27294	0.34977	0.58151	0.31703	0.72492	0.47096	1				
	p	0.3102	0.3241	0.3451	0.2202	*0.0292*	0.2694	*0.0034*	**0.0892**					
	n	14	14	14	14	14	14	14	14	14				
logsurftowat	r	-0.35282	0.2661	-0.02039	0.59187	-0.00005	-0.1211	-0.20983	-0.41864	0.52138	1			
	p	**0.216**	0.3578	0.9448	*0.0258*	0.9999	0.6801	0.4715	**0.1363**	**0.0559**				
	n	14	14	14	14	14	14	14	14	14	14			
logflow	r	-0.14824	0.26635	-0.27058	0.35661	0.68151	0.31642	0.77997	0.51055	0.95418	0.38729	1		
	p	0.613	0.3573	0.3495	0.2107	*0.0073*	0.2704	*0.001*	**0.0621**	*<.0001*	**0.1713**			
	n	14	14	14	14	14	14	14	14	14	14	14		
ResTime	r	0.1435	0.07537	-0.17439	-0.3684	-0.21726	0.5187	0.30332	0.46822	-0.18684	-0.63881	-0.217	1	
	p	0.6245	0.7979	0.551	0.195	0.4556	**0.0574**	0.2918	**0.0913**	0.5225	*0.0139*	0.4562		
	n	14	14	14	14	14	14	14	14	14	14	14	14	
logHgdepDNR	r	-0.271	0.19704	-0.40045	0.28095	0.72321	0.50583	0.72059	0.71665	0.68476	0.07609	0.73201	0.00487	1
	p	0.3487	0.4996	**0.1559**	0.3306	*0.0035*	**0.065**	*0.0036*	*0.0039*	*0.0069*	0.796	*0.0029*	0.9868	
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logLUdev	r	0.21129	-0.22403	0.03662	-0.49605	-0.19449	0.04206	-0.05074	0.04301	-0.49603	-0.64189	-0.40029	0.56505	-0.15241
	p	0.4684	0.4413	0.9011	**0.0712**	0.5052	0.8865	0.8632	0.8839	**0.0712**	*0.0133*	**0.1561**	*0.0352*	0.603
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logLUag	r	-0.13329	0.02608	-0.24617	0.28379	0.36841	0.2912	0.48519	0.22701	0.76516	0.48601	0.68509	-0.11472	0.50747
	p	0.6496	0.9295	0.3962	0.3255	**0.1949**	0.3124	**0.0786**	0.4351	*0.0014*	**0.0781**	*0.0069*	0.6961	**0.064**
	n	14	14	14	14	14	14	14	14	14	14	14	14	14

		logwatper MeHg	sedper MeHg	LMB	log LMBBAF	Age	Depth	logsurfarea	logcap	logwatarea	logsurftowat	logflow	ResTime	log HgdepDNR
logLUfor	r	0.31886	0.20934	0.30499	0.26289	0.29385	0.12345	0.19413	0.30922	0.15017	-0.02534	0.18271	-0.16699	0.10695
	p	0.2665	0.4726	0.289	0.3639	0.3079	0.6741	0.506	0.282	0.6084	0.9315	0.5318	0.5683	0.7159
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logLUwet	r	0.20111	-0.38219	0.23201	-0.12935	-0.20359	-0.5974	-0.30101	-0.6321	-0.10521	0.20789	-0.09876	-0.34786	-0.61498
	p	0.5532	0.2461	0.4924	0.7046	0.5482	0.0523	0.3684	0.0369	0.7582	0.5396	0.7727	0.2945	0.044
	n	11	11	11	11	11	11	11	11	11	11	11	11	11
logHgD	r	-0.82727	-0.32176	-0.10384	-0.15183	-0.06216	-0.26912	-0.06819	-0.05248	0.02614	0.12087	-0.11555	-0.12589	-0.00159
	p	0.0003	0.2619	0.7239	0.6043	0.8328	0.3522	0.8168	0.8586	0.9293	0.6806	0.694	0.6681	0.9957
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logHgUNF	r	-0.72613	-0.32796	-0.04762	-0.22861	-0.08574	-0.31664	-0.0471	-0.04931	0.00814	0.06964	-0.12635	-0.15065	-0.06678
	p	0.0033	0.2523	0.8716	0.4318	0.7707	0.27	0.873	0.8671	0.978	0.813	0.6669	0.6072	0.8206
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logMeHgD	r	0.65506	-0.23858	0.7498	-0.54624	-0.12566	-0.34469	-0.1763	-0.16652	-0.48094	-0.46151	-0.41559	0.08029	-0.47841
	p	0.011	0.4114	0.002	0.0433	0.6686	0.2275	0.5466	0.5694	0.0817	0.0967	0.1394	0.785	0.0835
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logMeHgUNF	r	0.43515	-0.27815	0.61593	-0.74933	-0.0951	-0.38887	-0.04338	-0.0586	-0.35776	-0.45166	-0.35007	0.15823	-0.38848
	p	0.1199	0.3356	0.019	0.002	0.7464	0.1694	0.8829	0.8423	0.2092	0.105	0.2198	0.589	0.1699
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
pH	r	-0.39297	0.08163	-0.67055	0.45394	0.17625	0.33213	0.07921	0.04112	0.29146	0.31141	0.33151	-0.06799	0.51885
	p	0.1645	0.7815	0.0087	0.103	0.5467	0.246	0.7878	0.889	0.312	0.2785	0.2469	0.8174	0.0573
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logTSS	r	-0.23799	-0.00363	0.06886	-0.47287	0.14163	-0.12369	0.22049	0.26044	0.20534	0.01866	0.04594	0.03066	0.17897
	p	0.4126	0.9902	0.8151	0.0877	0.6291	0.6735	0.4488	0.3685	0.4813	0.9495	0.8761	0.9171	0.5404
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logDOC	r	0.02899	-0.21816	0.14407	-0.21438	-0.06452	-0.30936	0.01883	-0.14085	0.0294	0.01993	-0.01737	0.2423	-0.10317
	p	0.9216	0.4537	0.6232	0.4618	0.8265	0.2818	0.9491	0.631	0.9205	0.9461	0.953	0.4039	0.7256
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logCl	r	-0.62648	-0.19022	-0.16223	-0.12655	0.01468	-0.10658	-0.09393	-0.03995	-0.01139	0.09634	-0.07636	-0.13124	0.27869
	p	0.022	0.5336	0.5964	0.6804	0.962	0.7289	0.7602	0.8969	0.9705	0.7542	0.8042	0.6691	0.3565
	n	13	13	13	13	13	13	13	13	13	13	13	13	13
logNO3	r	-0.04982	0.13526	0.46801	-0.04456	-0.09758	-0.17092	-0.0551	-0.17883	0.26045	0.43418	0.08332	-0.05421	-0.05701
	p	0.8778	0.6751	0.1249	0.8906	0.7629	0.5953	0.8649	0.5782	0.4136	0.1585	0.7968	0.8671	0.8603
	n	12	12	12	12	12	12	12	12	12	12	12	12	12
logSO4	r	0.27883	0.18319	0.03726	0.27408	0.01899	0.1279	-0.11524	-0.04729	-0.15985	-0.08422	-0.03509	-0.20437	-0.20014
	p	0.3344	0.5307	0.8994	0.343	0.9486	0.663	0.6948	0.8724	0.5851	0.7747	0.9052	0.4834	0.4927
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logbottdo	r	0.01925	0.10833	-0.00089	0.61113	-0.32555	0.09148	-0.42083	-0.46643	-0.04843	0.45967	-0.01225	-0.40746	-0.22113
	p	0.9502	0.7246	0.9977	0.0265	0.2777	0.7663	0.1522	0.1081	0.8752	0.114	0.9683	0.167	0.4678
	n	13	13	13	13	13	13	13	13	13	13	13	13	13
logpwHg	r	0.04594	0.33348	0.12258	-0.4353	-0.24486	0.21106	0.253	0.40661	-0.13833	-0.55888	-0.20347	0.49418	-0.16475
	p	0.8997	0.3464	0.7359	0.2086	0.4954	0.5583	0.4806	0.2436	0.7031	0.0931	0.5729	0.1465	0.6492
	n	10	10	10	10	10	10	10	10	10	10	10	10	10

		logwatper MeHg	sedper MeHg	LMB	log LMBBAF	Age	Depth	logsurfarea	logcap	logwatarea	logsurf towat	logflow	ResTime	log HgdepDNR
logpwMeHg	r	-0.03919	0.80686	-0.25444	-0.36733	-0.25	0.51221	0.44859	0.46249	0.11282	-0.41177	0.07041	0.77538	-0.04294
	p	0.9144	*0.0048*	0.4781	0.2964	0.486	0.1301	0.1935	0.1783	0.7563	0.2371	0.8467	*0.0084*	0.9062
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
log2pwFe	r	0.47995	0.21864	0.78241	-0.5395	-0.40821	-0.17591	-0.19694	-0.00919	-0.52785	-0.58987	-0.62346	0.47159	-0.64738
	p	0.1604	0.5439	*0.0075*	0.1075	0.2415	0.6269	0.5855	0.9799	0.1168	0.0727	0.0541	0.1688	*0.043*
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
logpwMn	r	-0.03844	-0.23342	0.31484	0.0189	0.13664	-0.17406	-0.06351	0.10842	-0.14615	-0.15038	-0.24432	-0.15173	-0.16177
	p	0.916	0.5163	0.3756	0.9587	0.7066	0.6306	0.8616	0.7656	0.687	0.6784	0.4963	0.6756	0.6553
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
pwDOC	r	0.2624	0.09584	0.54695	-0.16345	-0.11194	0.04147	0.1219	0.20866	-0.09322	-0.30775	-0.18648	0.26105	-0.30403
	p	0.4639	0.7923	0.1018	0.6519	0.7582	0.9094	0.7373	0.5629	0.7978	0.387	0.606	0.4663	0.3931
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
Bulkden	r	-0.24942	0.21051	-0.21134	-0.03759	-0.16124	0.30801	0.40122	0.29859	0.35815	0.05062	0.2943	0.17993	0.03026
	p	0.4871	0.5594	0.5578	0.9179	0.6563	0.3866	0.2505	0.402	0.3095	0.8895	0.4091	0.6189	0.9339
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
Por	r	0.10108	-0.0574	-0.03783	0.10487	0.12926	-0.11065	-0.28182	-0.2223	-0.23826	-0.01563	-0.15628	-0.06003	0.07651
	p	0.7811	0.8748	0.9174	0.7731	0.7219	0.7609	0.4302	0.5371	0.5074	0.9658	0.6664	0.8692	0.8336
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
dw	r	-0.16201	0.03601	-0.00623	-0.1014	-0.1308	0.12321	0.29815	0.22461	0.27709	0.0569	0.18892	0.03821	-0.06252
	p	0.6548	0.9213	0.9864	0.7805	0.7187	0.7345	0.4027	0.5327	0.4383	0.8759	0.6012	0.9165	0.8638
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
LOI	r	0.02438	-0.0422	-0.08157	0.05175	0.00391	-0.15502	-0.09871	-0.39033	0.22822	0.44569	0.22407	-0.16498	-0.01072
	p	0.9341	0.8861	0.7816	0.8605	0.9894	0.5967	0.7371	0.1676	0.4326	0.1102	0.4412	0.573	0.971
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logFeII	r	-0.14156	-0.44452	0.02073	-0.15204	0.40721	0.02854	0.00896	0.2013	-0.13578	-0.23643	-0.10055	-0.11451	0.3533
	p	0.6965	0.198	0.9547	0.675	0.2428	0.9376	0.9804	0.5771	0.7084	0.5108	0.7822	0.7528	0.3166
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
logFeIII	r	0.26821	-0.29005	0.65111	-0.12923	-0.07221	-0.40705	-0.18263	-0.13923	-0.17528	-0.03717	-0.25991	-0.2016	-0.34197
	p	0.4537	0.4163	*0.0414*	0.722	0.8429	0.243	0.6136	0.7013	0.6281	0.9188	0.4683	0.5765	0.3335
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
logsedHg	r	-0.14429	-0.18315	0.02988	-0.27151	0.21876	-0.14847	0.15204	-0.00427	0.20084	0.09521	0.19006	-0.00137	0.25745
	p	0.6226	0.5308	0.9192	0.3477	0.4524	0.6125	0.6038	0.9884	0.4911	0.7461	0.5152	0.9963	0.3742
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logsedMeHg	r	-0.08026	0.48645	0.18391	0.04822	0.16886	0.16183	0.17214	0.14174	0.3173	0.23618	0.28577	0.02384	0.2977
	p	0.7851	0.0778	0.5291	0.87	0.5639	0.5805	0.5562	0.6288	0.269	0.4163	0.322	0.9355	0.3013
	n	14	14	14	14	14	14	14	14	14	14	14	14	14
logAVS	r	0.27025	-0.32555	0.41648	-0.56677	-0.1118	-0.39897	-0.37497	-0.1643	-0.52979	-0.36282	-0.5505	-0.07566	-0.30613
	p	0.4501	0.3587	0.2312	0.0876	0.7585	0.2534	0.2857	0.6501	0.1152	0.3028	0.0991	0.8354	0.3896
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
logCRS	r	0.44978	-0.23049	0.66839	-0.53873	-0.07771	-0.49577	-0.39898	-0.23093	-0.53982	-0.34533	-0.56312	-0.0122	-0.34619
	p	0.1921	0.5218	*0.0346*	0.1081	0.831	0.145	0.2534	0.5209	0.1073	0.3284	0.0901	0.9733	0.3271
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
logkdHg	r	-0.03476	-0.14202	-0.17269	0.20815	0.27999	-0.00456	-0.03996	-0.17882	0.19348	0.36471	0.26675	-0.2115	0.29529
	p	0.9241	0.6955	0.6333	0.5639	0.4333	0.99	0.9127	0.6211	0.5923	0.3001	0.4563	0.5575	0.4075
	n	10	10	10	10	10	10	10	10	10	10	10	10	10
logkdMeHg	r	0.02368	-0.53591	0.05743	0.23683	0.12615	-0.43571	-0.41191	-0.52926	-0.02414	0.50644	0.02776	-0.61348	0.02153
	p	0.9482	0.1103	0.8748	0.51	0.7284	0.2081	0.2369	0.1157	0.9472	0.1353	0.9393	0.0592	0.9529
	n	10	10	10	10	10	10	10	10	10	10	10	10	10

		logLUdev	logLUag	logLUfor	logLUwet	logHgD	logHgUNF	logMeHgD	HgUNF	pH	logTSS	logDOC	logCl	logNO3	logSO4
logLUdev	r	1													
	p														
	n	14													
logLUag	r	-0.57945	1												
	p	*0.0299*													
	n	14	14												
logLUfor	r	-0.45511	0.05937	1											
	p	**0.102**	0.8402												
	n	14	14	14											
logLUwet	r	-0.23731	0.19136	-0.11439	1										
	p	0.4823	0.573	0.7377											
	n	11	11	11	11										
logHgD	r	0.00833	-0.06702	-0.35893	-0.05515	1									
	p	0.9774	0.8199	**0.2076**	0.8721										
	n	14	14	14	11	14									
logHgUNF	r	-0.13998	-0.00047	-0.29808	0.08819	0.94504	1								
	p	0.6331	0.9987	0.3006	0.7965	*<.0001*									
	n	14	14	14	11	14	14								
logMeHgD	r	0.3834	-0.32268	0.07073	0.30584	-0.11762	-0.01034	1							
	p	**0.176**	0.2605	0.8101	0.3604	0.6888	0.972								
	n	14	14	14	11	14	14	14							
logMeHgUNF	r	0.44079	-0.32726	-0.08685	0.23539	0.09977	0.18752	0.90401	1						
	p	**0.1147**	0.2534	0.7678	0.4859	0.7344	0.5209	*<.0001*							
	n	14	14	14	11	14	14	14	14						
pH	r	0.08068	0.18149	-0.30654	-0.33912	-0.05523	-0.27307	-0.76754	-0.74691	1					
	p	0.7839	0.5346	0.2864	0.3076	0.8512	0.3449	*0.0014*	*0.0021*						
	n	14	14	14	11	14	14	14	14	14					
logTSS	r	-0.14551	0.12792	-0.09518	-0.12386	0.38419	0.46221	0.09937	0.43946	-0.33058	1				
	p	0.6196	0.663	0.7462	0.7167	**0.175**	**0.0961**	0.7354	**0.1159**	**0.2483**					
	n	14	14	14	11	14	14	14	14	14	14				
logDOC	r	0.34602	-0.03685	-0.26767	0.06128	0.08686	0.04545	0.16544	0.30754	-0.10167	0.0761	1			
	p	**0.2256**	0.9005	0.3549	0.8579	0.7678	0.8774	0.5719	0.2848	0.7295	0.796				
	n	14	14	14	11	14	14	14	14	14	14	14			
logCl	r	0.25945	-0.06429	-0.62922	-0.41376	0.74406	0.666	-0.10398	0.08655	0.22782	0.40633	0.15755	1		
	p	0.392	0.8347	*0.0212*	**0.2346**	*0.0035*	*0.0129*	0.7353	0.7786	0.4541	**0.1683**	0.6072			
	n	13	13	13	10	13	13	13	13	13	13	13	13		
logNO3	r	-0.1781	0.33152	-0.17161	0.10116	0.39739	0.48797	0.27557	0.38254	-0.35992	0.4966	0.66979	0.3273	1	
	p	0.5797	0.2925	0.5938	0.7957	**0.2008**	**0.1075**	0.386	**0.2197**	0.2505	**0.1005**	*0.0172*	0.299		
	n	12	12	12	9	12	12	12	12	12	12	12	12	12	
logSO4	r	-0.10067	-0.19498	0.30786	0.13554	-0.36084	-0.38106	0.0081	-0.24555	0.08288	-0.51268	-0.76738	-0.4929	-0.72232	1
	p	0.732	0.5041	0.2843	0.6911	**0.205**	**0.1789**	0.9781	0.3975	0.7782	**0.0609**	*0.0014*	**0.087**	*0.008*	
	n	14	14	14	11	14	14	14	14	14	14	14	13	12	14
logbottdo	r	-0.19557	-0.02474	0.29147	0.17566	-0.19996	-0.31967	-0.24208	-0.51021	0.34039	-0.75391	-0.19752	-0.3678	-0.26655	0.45881
	p	0.522	0.9361	0.3339	0.6274	0.5125	0.287	0.4255	**0.0748**	0.2551	*0.0029*	0.5177	**0.2394**	0.4282	**0.1148**
	n	13	13	13	10	13	13	13	13	13	13	13	12	11	13

		logLUdev	logLUag	logLUfor	logLUwet	logHgD	logHgUNF	logMeHgD	HgUNF	pH	logTSS	logDOC	logCl	logNO3	logSO4
logpwHg	r	-0.13036	-0.15526	0.39964	-0.07832	0.11441	0.38092	0.22378	0.3256	-0.69627	0.36061	0.08028	-0.22516	0.37071	-0.34375
	p	0.7196	0.6684	0.2525	0.8538	0.753	0.2775	0.5343	0.3586	0.0253	0.306	0.8255	0.5603	0.366	0.3308
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
logpwMeHg	r	0.18376	-0.11118	-0.2038	-0.29498	-0.04735	0.05648	-0.13051	0.09287	-0.2135	0.26932	0.33546	-0.19055	0.21821	-0.34947
	p	0.6113	0.7598	0.5722	0.4782	0.8967	0.8768	0.7193	0.7986	0.5537	0.4518	0.3433	0.6234	0.6037	0.3223
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
log2pwFe	r	0.32117	-0.4577	0.28881	0.20351	-0.04517	0.04774	0.8071	0.81559	-0.82533	0.174	0.04749	-0.52339	0.19897	0.18912
	p	0.3655	0.1835	0.4183	0.6288	0.9014	0.8958	0.0048	0.004	0.0033	0.6307	0.8963	0.1482	0.6367	0.6008
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
logpwMn	r	-0.52377	0.14292	0.5148	0.1872	0.24861	0.45075	0.24382	0.11962	-0.54505	0.23661	-0.77665	-0.13098	-0.00942	0.4986
	p	0.1202	0.6937	0.1279	0.6571	0.4886	0.1911	0.4972	0.742	0.1032	0.5104	0.0082	0.737	0.9823	0.1424
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
pwDOC	r	-0.33707	0.12102	0.74126	0.24579	0.0176	0.26158	0.49142	0.39826	-0.85067	0.1525	-0.08701	-0.57339	0.5486	0.00567
	p	0.3409	0.7391	0.0142	0.5574	0.9615	0.4654	0.1492	0.2543	0.0018	0.6741	0.8111	0.1065	0.1591	0.9876
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
Bulkden	r	-0.5405	0.3603	0.39119	0.1653	0.20806	0.45684	-0.19661	-0.13276	-0.40137	0.18502	0.11226	-0.24358	0.72421	-0.4236
	p	0.1067	0.3064	0.2636	0.6957	0.5641	0.1844	0.5862	0.7147	0.2503	0.6088	0.7575	0.5277	0.0422	0.2225
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
Por	r	0.62884	-0.34419	-0.59753	-0.27591	-0.17224	-0.46619	-0.02732	-0.05067	0.60583	-0.27215	0.02385	0.35092	-0.78677	0.32134
	p	0.0515	0.3301	0.0681	0.5083	0.6342	0.1744	0.9403	0.8894	0.0634	0.4468	0.9479	0.3545	0.0205	0.3653
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
dw	r	-0.64328	0.36366	0.57228	0.27281	0.22766	0.50985	-0.01397	0.02323	-0.57561	0.28324	-0.05101	-0.31298	0.81103	-0.30659
	p	0.0448	0.3016	0.0838	0.5133	0.527	0.1322	0.9695	0.9492	0.0816	0.4278	0.8887	0.4122	0.0146	0.3889
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
LOI	r	0.26138	0.16528	-0.52907	0.26263	-0.11033	-0.28695	-0.10395	-0.02101	0.48925	-0.09294	0.29448	0.11106	0.20181	-0.0601
	p	0.3667	0.5723	0.0517	0.4353	0.7073	0.3199	0.7236	0.9432	0.0758	0.752	0.3068	0.7179	0.5294	0.8383
	n	14	14	14	11	14	14	14	14	14	14	14	13	12	14
logFeII	r	0.46898	-0.31449	-0.04803	-0.66999	0.27308	0.01246	0.08619	0.14623	0.33437	0.12514	-0.23923	0.63176	-0.59262	0.17164
	p	0.1715	0.3761	0.8952	0.0691	0.4452	0.9727	0.8129	0.6869	0.345	0.7305	0.5056	0.068	0.1216	0.6354
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
logFeIII	r	-0.47023	0.13152	0.75958	0.53882	0.04489	0.27836	0.53399	0.44018	-0.77033	0.23806	-0.19523	-0.46165	0.53141	-0.02148
	p	0.1702	0.7172	0.0108	0.1682	0.902	0.4361	0.1119	0.203	0.0091	0.5078	0.5888	0.211	0.1753	0.953
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
logsedHg	r	0.52435	0.00914	-0.5747	-0.20267	0.28691	0.1107	0.1349	0.34951	0.27001	0.28736	0.48352	0.6159	0.41216	-0.43987
	p	0.0542	0.9753	0.0316	0.5501	0.32	0.7064	0.6457	0.2206	0.3505	0.3192	0.0798	0.025	0.1831	0.1155
	n	14	14	14	11	14	14	14	14	14	14	14	13	12	14
logsedMeHg	r	0.27802	0.00918	-0.45651	-0.54936	0.12081	0.01112	0.02528	0.19611	0.17986	0.33125	0.24763	0.49272	0.49631	-0.28247
	p	0.3358	0.9751	0.1008	0.08	0.6808	0.9699	0.9316	0.5016	0.5384	0.2473	0.3933	0.0871	0.1008	0.3278
	n	14	14	14	11	14	14	14	14	14	14	14	13	12	14
logAVS	r	0.54459	-0.63478	-0.13547	-0.25786	0.05303	-0.08525	0.56164	0.63315	-0.15852	0.22688	-0.25068	0.2115	-0.44959	0.44851
	p	0.1036	0.0486	0.709	0.5375	0.8843	0.8149	0.0911	0.0494	0.6618	0.5285	0.4848	0.5849	0.2637	0.1936
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
logCRS	r	0.54192	-0.56067	0.03172	0.03909	-0.08894	-0.20452	0.70056	0.78161	-0.27727	0.27845	-0.00596	-0.04293	-0.20441	0.16397
	p	0.1056	0.0918	0.9307	0.9268	0.807	0.5709	0.024	0.0076	0.438	0.436	0.987	0.9127	0.6273	0.6508
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
logkdHg	r	0.40565	0.03464	-0.44916	-0.15834	-0.12297	-0.44989	-0.21747	-0.17964	0.73284	-0.19826	0.20196	0.29203	-0.34473	0.04057
	p	0.2448	0.9243	0.1928	0.708	0.735	0.192	0.5461	0.6195	0.0159	0.5829	0.5758	0.4458	0.403	0.9114
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10
logkdMeHg	r	0.16065	-0.00338	-0.2878	0.16524	-0.04591	-0.28915	-0.01373	-0.09021	0.51676	-0.25247	-0.07842	0.24847	-0.31783	0.29841
	p	0.6575	0.9926	0.42	0.6958	0.8998	0.4178	0.97	0.8043	0.1262	0.4816	0.8295	0.5191	0.443	0.4023
	n	10	10	10	8	10	10	10	10	10	10	10	9	8	10

		logbottdo	logpwHg	logpw MeHg	log2pwFe	logpwMn	pwDOC	Bulkden	Por	dw	LOI	logFeII	logFeIII	logsedHg	logsed MeHg
logbottdo	r	1													
	p														
	n	13													
logpwHg	r	-0.52993	1												
	p	0.1151													
	n	10	10												
logpwMeHg	r	-0.57818	0.68399	1											
	p	0.08	0.0292												
	n	10	10	10											
log2pwFe	r	-0.25393	0.51741	0.30568	1										
	p	0.479	0.1256	0.3904											
	n	10	10	10	10										
logpwMn	r	-0.19738	0.3612	-0.16863	0.36384	1									
	p	0.5847	0.3051	0.6414	0.3014										
	n	10	10	10	10	10									
pwDOC	r	-0.16055	0.73937	0.22943	0.67775	0.67651	1								
	p	0.6577	*0.0145*	0.5237	*0.0313*	*0.0317*									
	n	10	10	10	10	10	10								
Bulkden	r	-0.13568	0.76879	0.52808	0.07975	0.24276	0.6275	1							
	p	0.7086	*0.0094*	0.1166	0.8267	0.4992	0.0521								
	n	10	10	10	10	10	10	10							
Por	r	0.15339	-0.78842	-0.35778	-0.23474	-0.46284	-0.79291	-0.94662	1						
	p	0.6722	*0.0067*	0.3101	0.5139	0.178	*0.0062*	*<.0001*							
	n	10	10	10	10	10	10	10	10						
dw	r	-0.13514	0.76473	0.35191	0.20693	0.46023	0.76927	0.95242	-0.99691	1					
	p	0.7097	*0.01*	0.3187	0.5662	0.1808	*0.0093*	*<.0001*	*<.0001*						
	n	10	10	10	10	10	10	10	10	10					
LOI	r	0.18863	-0.80187	-0.22288	-0.3127	-0.65061	-0.70192	-0.57333	0.66877	-0.64502	1				
	p	0.5371	*0.0053*	0.5359	0.379	*0.0416*	*0.0237*	0.0831	*0.0345*	*0.044*					
	n	13	10	10	10	10	10	10	10	10	14				
logFeII	r	-0.05214	-0.48936	-0.49252	-0.13384	-0.05391	-0.43887	-0.70567	0.65737	-0.64062	0.15104	1			
	p	0.8863	0.1511	0.1481	0.7124	0.8824	0.2045	*0.0226*	*0.0389*	*0.046*	0.677				
	n	10	10	10	10	10	10	10	10	10	10	10			
logFeIII	r	0.01116	0.45027	-0.20502	0.52226	0.65242	0.81878	0.40774	-0.66723	0.64377	-0.51129	-0.31048	1		
	p	0.9756	0.1916	0.5699	0.1215	*0.0409*	*0.0038*	0.2421	*0.0351*	*0.0446*	0.1309	0.3826		0.479307	
	n	10	10	10	10	10	10	10	10	10	10	10	10		
logsedHg	r	-0.28488	-0.64133	-0.16437	-0.24703	-0.68881	-0.71877	-0.64296	0.74645	-0.72092	0.69232	0.6525	-0.61633	1	
	p	0.3455	*0.0457*	0.65	0.4914	*0.0276*	*0.0192*	*0.0449*	*0.0131*	*0.0186*	*0.0061*	*0.0409*	0.0577		
	n	13	10	10	10	10	10	10	10	10	14	10	10	14	
logsedMeHg	r	-0.29452	-0.40376	0.2614	-0.1153	-0.68561	-0.65508	-0.51815	0.68566	-0.6679	0.50092	0.42865	-0.76397	0.737	1
	p	0.3287	0.2472	0.4657	0.7511	*0.0286*	*0.0398*	0.125	*0.0286*	*0.0348*	0.0681	0.2165	*0.0101*	*0.0026*	
	n	13	10	10	10	10	10	10	10	10	14	10	10	14	14
logAVS	r	-0.16673	-0.29314	-0.28689	0.37683	0.06717	-0.22546	-0.73098	0.59882	-0.59764	0.18941	0.71075	-0.10835	0.47278	0.3965
	p	0.6452	0.4111	0.4216	0.2831	0.8537	0.5311	*0.0163*	*0.0674*	0.068	0.6002	*0.0212*	0.7657	0.1676	0.2566
	n	10	10	10	10	10	10	10	10	10	10	10	10	10	10
logCRS	r	-0.14784	-0.22913	-0.30442	0.52821	-0.01667	-0.07195	-0.66397	0.48015	-0.49833	0.24127	0.54306	0.17941	0.44777	0.31591
	p	0.6836	0.5243	0.3924	0.1165	0.9635	0.8434	*0.0363*	0.1602	0.1427	0.5019	0.1048	0.6199	0.1944	0.3739
	n	10	10	10	10	10	10	10	10	10	10	10	10	10	10
logkdHg	r	0.32666	-0.90372	-0.44716	-0.45343	-0.62964	-0.81834	-0.76665	0.847	-0.82514	0.85927	0.58214	-0.5997	0.89407	0.70605
	p	0.3569	*0.0003*	0.1951	0.1881	0.0511	*0.0038*	*0.0097*	*0.002*	*0.0033*	*0.0014*	0.0775	0.0669	*0.0005*	*0.0225*
	n	10	10	10	10	10	10	10	10	10	10	10	10	10	10
logkdMeHg	r	0.54254	-0.94667	-0.78919	-0.38553	-0.32227	-0.66651	-0.78611	0.74054	-0.72034	0.74187	0.55255	-0.26322	0.64195	0.31757
	p	0.1052	*<.0001*	*0.0066*	0.2712	0.3638	*0.0353*	*0.007*	*0.0143*	*0.0188*	*0.014*	0.0976	0.4625	*0.0454*	0.3712
	n	10	10	10	10	10	10	10	10	10	10	10	10	10	10

		logAVS	logCRS	logkdHg	logkd MeHg
logAVS	r	1			
	p				
	n	10			
logCRS	r	0.85049	1		
	p	0.0018			
	n	10	10		
logkdHg	r	0.34593	0.33752	1	
	p	0.3275	0.3402		
	n	10	10	10	
logkdMeHg	r	0.44107	0.43554	0.8601	1
	p	0.202	0.2083	0.0014	
	n	10	10	10	10